

Newtonian Kinetic Theory and the Ergodic-Nonergodic Transition

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Abstract

In a recent work we have discussed how kinetic theory, the statistics of classical particles obeying Newtonian dynamics, can be formulated as a field theory. The field theory can be organized to produce a self-consistent perturbation theory expansion in an effective interaction potential. In the present work we use this development for investigating ergodic-nonergodic (ENE) transitions in dense fluids. The theory is developed in terms of a core problem spanned by the variables ρ , the number density, and B , a response density. We set up the perturbation theory expansion for studying the self-consistent model which gives rise to a ENE transition. Our main result is that the low-frequency dynamics near the ENE transition is the same for Smoluchowski and Newtonian dynamics. This is true despite the fact that term by term in a density expansion the results for the two dynamics are fundamentally different.

I. INTRODUCTION

In previous work [1], referred to as ND1 from hereon, we showed how the Newtonian dynamics (ND) of simple fluids can be treated using field-theoretic methods[2]. Here we apply these methods to systems in equilibrium with an emphasis on the ergodic-nonergodic (ENE) transition which is generally associated with the mode coupling models[3–5] for glassy dynamics. We analyze the long-time behavior of the density-density time correlation function. For high densities we find that there is a slow dynamical regime which is very similar to that found in the case of Smolukowski dynamics (SD)[6, 7]. In the shorter-time regimes the SD dynamics is considerably simpler than ND. The complications in the ND case arise because of the unwieldy forms of the three-point vertices even for the ideal-gas limit. In the long-time limit we find that the three-point cumulants and three-point vertices satisfy fluctuation-dissipation relations which are very helpful in treating the late-stage kinetics at high densities.

After determining the nature of the fluctuation dissipation relations[8] for the two-point cumulants and vertices, we derive an exact expression (kinetic equation) satisfied by the density-density correlation function. The form of this kinetic equation is the same as found in projection operator treatments[9] of the problem. This approach involves the introduction of memory functions. The significant difference between our work here and the projection operator method is that we obtain expressions for the memory functions which are suitable for perturbation theory analysis. It is not expressed in term of "projected dynamics". Instead the memory function is expressed as the "BB" component of a two-point vertex function Γ_{BB} associated with what we call a core problem. The core problem involves the kinetics of the particle density ρ and a response field "B".

We then carry out perturbation theory in terms of a pseudo potential and find at second order that the memory function kernel K_{BB} in the low-frequency regime is proportional to a quadratic-form in the full density-density correlation function. This suggests a non-linear feedback loop similar to that found in mode-coupling theory[10, 11] and in treating SD[6].

When we focus on long-time solutions we find that the developments for SD and ND are the same. In SDENE[6] we analyzed the leading contribution near an ENE transition. In SM[12] it is shown the ENE-transition is accompanied with a two-step decay process. SM shows that there are a pair of exponents, a and b , which characterize the slow-time power-

law solutions. They treat in detail the case of hard spheres at second order in perturbation theory. We find here that SD and ND systems share the same ENE transition according to our results at second order in perturbation theory. This point is amplified in Ref.13 where higher order terms in perturbation theory are treated. We speculate that there is a whole class of systems which share the same statics and ENE transition.

II. FIELD-THEORETICAL FORMULATION

A. Newtonian Equations of Motion

We discuss the kinetics satisfied by a system of N particles with mass m , position R_i and momentum P_i we consider Newtonian dynamics, with equations of motion

$$m\dot{R}_i = P_i \quad (2.1)$$

$$\dot{P}_i = f_i \quad , \quad (2.2)$$

where the force is given by

$$f_i = -\nabla_i U \quad , \quad (2.3)$$

the total potential energy is

$$U = \frac{1}{2} \sum_{i,j} V(R_i - R_j) \quad (2.4)$$

where $V(R_i - R_j)$ is a standard pair potential between the i -th and j -th particles. Starting with these equations of motion we can reexpress the problem in terms of a path-integral formulation.

B. Path-Integral Formulation

In FTSPD and ND1 we introduced a field-theoretical formulation for classical many-particles dynamics. Here we summarize the results for Newtonian dynamics. The grand partition function for a *core* set of dynamical fields $\{\Phi_\alpha\} \equiv \Phi$ is given by

$$Z_T = \sum_{N=0}^{\infty} \frac{z^N}{N!} \text{Tr} e^{-\mathcal{A} + H \cdot \Phi} \quad (2.5)$$

where the trace is over the set of phase-space variables $R_i(t)$ and $P_i(t)$, and the conjugate MSR[14] variables $\hat{R}_i(t)$ and $\hat{P}_i(t)$. z is the fugacity and H_i is an external time-dependent field that couples to the set of dynamical variables Φ_α . The MSR action \mathcal{A} is given by

$$\mathcal{A} = \sum_i \int \left[i\hat{R}_i(t) \left\{ \dot{R}_i(t) - \frac{P_i(t)}{m} \right\} + i\hat{P}_i(t) \left\{ \dot{P}_i(t) - f_i(t) \right\} \right] dt \quad . \quad (2.6)$$

In the present work the set $\{\Phi_\alpha\}$ stands for the core variables $\{\rho, B\}$ which are collective properties and are expressed as a sum of one point functions. Explicitly the particle density is given by

$$\rho(1) = \sum_{i=1}^N \delta(x_1 - R_i(t_1)), \quad (2.7)$$

and the response field B :

$$B(x, t) = i \sum_{i=1}^N \hat{P}_i \cdot \frac{\partial}{\partial R_i} \delta(x - R_i(t)) \quad . \quad (2.8)$$

The B -field is important in the present analysis and is somewhat unfamiliar. We note that B is the longitudinal component of the vector field $\hat{\mathbf{g}}(x, t)$. The latter is expressed as a collective density corresponding to the hatted MSR field $\hat{\mathbf{P}}_i$ (with a factor i) conjugate to the momentum \mathbf{P}_i of the i -th particle.

$$\hat{\mathbf{g}}(x, t) = \sum_{i=1}^N i\hat{\mathbf{P}}_i \delta(x - R_i(t)) \quad (2.9)$$

This $\hat{\mathbf{g}}(x, t)$ however is not the field conjugate to momentum density $\mathbf{g}(x, t)$ used in the usual MSR formulation of equations of nonlinear fluctuating hydrodynamics. The hatted fields in the MSR field theory are generally viewed as convenient mathematical tools for ensuring that the stochastic equations of the dynamics are satisfied in the field theory. These auxiliary fields give rise in a natural way to a set response functions which are related to correlation functions through suitable fluctuation dissipation relations. From such considerations, the B is best interpreted here as a suitable mean to obtain linear fluctuation dissipation relations facilitating the analysis of the dynamics developed here.

In general it is assumed that we can express Φ_α as a sum of one particle contributions

$$\Phi_\alpha = \sum_{i=1}^N \phi_\alpha^{(i)} \quad , \quad (2.10)$$

where with the roman index i we indicate that the corresponding quantity is a single-particle property. In the above equation we have used a compact notation where the index α labels space, time and fields ρ or B . We maintain the notation from here on. For $\alpha \equiv \rho$ we obtain the collective density for which we have

$$\phi_\rho^{(i)}(1) = \delta(x_1 - R_i(t_1)) \quad . \quad (2.11)$$

For $\alpha \equiv B$ field, the corresponding $\phi_B^{(i)}(1)$ is strongly dependent on the type of dynamics. For the SD case[2]

$$\phi_B^{(i)}(1) = D[i\hat{R}_i(t_1)\nabla_{x_1} + \frac{1}{2}\nabla_{x_1}^2]\delta(x_1 - R_i(t_1)) \quad (2.12)$$

while for the ND[1] case the corresponding result is

$$\phi_B^{(i)}(1) = -i\hat{P}_i(t_1)\nabla_{x_1}\delta(x_1 - R_i(t_1)) \quad (2.13)$$

where $\hat{P}_i(t_1)$ is the MSR conjugate momentum for the selected particle.

For our purposes the MSR action \mathcal{A} defined in eqn. (2.6) can be written as a sum of two parts,

$$\mathcal{A} = \mathcal{A}_0 + \mathcal{A}_I \quad . \quad (2.14)$$

The first term on the RHS \mathcal{A}_0 is the noninteracting MSR action. The interacting part of the action is given by

$$\mathcal{A}_I = \frac{1}{2} \sum_{\alpha, \nu} \int d1 d2 \Phi_\alpha(1) \sigma_{\alpha\nu}(12) \Phi_\nu(2) \quad (2.15)$$

where the Greek labels range over ρ and B . The interaction matrix, for systems in equilibrium in the distant past, is defined just as in SD, by

$$\sigma_{\alpha\beta}(12) = V(12) [\delta_{\alpha B} \delta_{\beta \rho} + \delta_{\alpha \rho} \delta_{\beta B}] \quad (2.16)$$

and

$$V(12) = V(x_1 - x_2)\delta(t_1 - t_2) \quad . \quad (2.17)$$

Notice that the response field B is chosen such that the interaction part of the action has the form given by Eq.(2.15).

For studying the dynamics of a many-particle system, we will be interested in the cumulants generated by corresponding generating functional

$$W[H] = \ln Z_T[H] \quad . \quad (2.18)$$

The one-point average of a field Φ_α defined in eqn. (2.10) in an external field is obtained in terms of the functional derivative

$$G_\alpha = \langle \Phi_\alpha \rangle = \frac{\delta}{\delta H_\alpha} W[H] \quad . \quad (2.19)$$

The full cumulants $G_{\alpha\beta}$, $G_{\alpha\beta\gamma}$ etc. are defined by successive functional derivatives of G_α ,

$$G_{\alpha\beta} = \frac{\delta}{\delta H_\beta} G_\alpha \quad (2.20)$$

$$G_{\alpha\beta\gamma} = \frac{\delta}{\delta H_\gamma} G_{\alpha\beta} \quad (2.21)$$

$$G_{\alpha\beta\dots\delta} = \frac{\delta}{\delta H_\alpha} \frac{\delta}{\delta H_\beta} \dots \frac{\delta}{\delta H_\delta} W[H] \quad . \quad (2.22)$$

We also need to deal with the *single-particle*[15] quantities

$$\mathcal{G}_{\alpha\beta\dots\delta} = \left\langle \sum_{i=1}^N \phi_\alpha^{(i)} \phi_\beta^{(i)} \dots \phi_\delta^{(i)} \right\rangle \quad . \quad (2.23)$$

III. FLUCTUATION DISSIPATION RELATIONS

In this section we explore the nature of fluctuation-dissipation relations in case of Newtonian dynamics assumed to be in equilibrium. The correlation functions between the fields in the field theory is primarily determined with the corresponding action functional of the associated field theory. The MSR action without initial conditions and external fields is given by Eq. (2.6). We now explore the invariance of the action \mathcal{A} under certain symmetry operations. In a stationary state we have time-translational invariance and time-reversal symmetry.

A. Symmetry transformations

1. The time-reversal symmetry T is defined as,

$$\begin{aligned} TR_i(t) &= R_i(-t) \\ T\hat{R}_i(t) &= -\hat{R}_i(-t) \\ TP_i(t) &= -P_i(-t) \\ T\hat{P}_i(t) &= \hat{P}_i(-t) \quad . \end{aligned} \tag{3.1}$$

We consider how the MSR-action \mathcal{A} changes under time-reversal

$$\begin{aligned} \mathcal{A}' = T\mathcal{A} &= \sum_i \int dt \left[i \left\{ -\hat{R}_i(-t) \right\} \left\{ -\frac{\partial R_i(-t)}{\partial(-t)} + \frac{P_i(-t)}{m} \right\} \right. \\ &\quad \left. + i \left\{ \hat{P}_i(-t) \right\} \left\{ \frac{\partial P_i(-t)}{\partial(-t)} - f_i(-t) \right\} \right] \end{aligned} \tag{3.2}$$

Letting $t \rightarrow -t$ in the time integral, we obtain $\mathcal{A}' = T\mathcal{A} = \mathcal{A}$ if the limits of integration are symmetric. This includes $t_2 \rightarrow \infty$ and $t_1 \rightarrow -\infty$ here. We conclude that the MSR action remains invariant under time-reversal.

2. In ND1 we have discussed the Fluctuation-Dissipation symmetry (FDS) for the field theory corresponding to the MSR action functional defined in Eq. (2.6). It was demonstrated [16] that the following transformation τ keeps the MSR action invariant :

$$\tau R_i(t) = R_i(-t) \tag{3.3}$$

$$\tau \hat{R}_i(t) = -\hat{R}_i(-t) - i\beta f_i(-t) \tag{3.4}$$

$$\tau P_i(t) = -P_i(-t) \tag{3.5}$$

$$\tau \hat{P}_i(t) = \hat{P}_i(-t) - i\frac{\beta}{m} P_i(-t) \quad . \tag{3.6}$$

We have therefore $\tau\mathcal{A} = \mathcal{A}$.

3. The inversion symmetry denoted by τ_1 is defined as

$$\begin{aligned}
\tau_1 R_i(t) &= -R_i(t) \\
\tau_1 \hat{R}_i(t) &= -\hat{R}_i(t) \\
\tau_1 P_i(t) &= -P_i(t) \\
\tau_1 \hat{P}_i(t) &= -\hat{P}_i(t) \quad .
\end{aligned} \tag{3.7}$$

The following invariant properties of the Action \mathcal{A} easily follows : $\tau_1 \mathcal{A} = \mathcal{A}$.

4. Finally we define the symmetry τ_2 as

$$\begin{aligned}
\tau_2 R_i(t) &= R_i(t) \\
\tau_2 \hat{R}_i(t) &= -\hat{R}_i(t) \\
\tau_2 P_i(t) &= P_i(t) \\
\tau_2 \hat{P}_i(t) &= -\hat{P}_i(t) \quad .
\end{aligned} \tag{3.8}$$

The following invariant properties of the Action $\tau_2 \mathcal{A}^* = \mathcal{A}$ is satisfied.

B. Fluctuation-Dissipation symmetry

From the definitions (2.7) and (2.8) it follows that under the transformation τ the core fields $\rho(x, t)$ and $B(x, t)$ change as

$$\tau \rho(x, t) = \rho(x, -t) \tag{3.9}$$

$$\tau B(x, t) = B(x, -t) - \beta \frac{\partial \rho(x, -t)}{\partial t} \tag{3.10}$$

In ND1 we had demonstrated the following Fluctuation dissipation theorem involving two-point correlation functions between the ρ and B fields.

$$G_{fB}(t - t') = \theta(t - t') \beta \frac{\partial}{\partial t} G_{f\rho}(t - t'), \tag{3.11}$$

for any function $f[\rho]$.

Here we are interested in higher-order FDR which are most easily expressed in terms of Fourier transforms. Consider the mixed correlation function

$$C_{BB\dots B\rho\dots\rho}(12\dots\ell\ell+1\dots n) = \left\langle B(1)B(2)\dots B(\ell)\rho(\ell+1)\dots\rho(n) \right\rangle \quad . \quad (3.12)$$

where

$$\rho(j) = \rho(q_j, \omega_j) = \int dt e^{i\omega_j t} \sum_{i=1}^N e^{-iq_j \cdot R_i(t)} \quad (3.13)$$

and

$$B(j) = B(q_j, \omega_j) = \int dt e^{i\omega_j t} (-q_j) \cdot \sum_{i=1}^N \hat{P}_i(t) e^{-iq_j \cdot R_i(t)} \quad (3.14)$$

Under the transformation τ we have

$$\tau B(1) = B(q_1, -\omega_1) + i\beta\omega_1\rho(q_1, -\omega_1) \quad (3.15)$$

$$\tau\rho(1) = \rho(q_1, -\omega_1) \quad . \quad (3.16)$$

Hence the mixed correlation defined above in Eq. (3.12) is obtained as

$$\begin{aligned} C_{BB\dots B\rho\dots\rho}(12\dots\ell\ell+1\dots n) &= \left\langle \tau B(1)\tau B(2)\dots\tau B(\ell)\tau\rho(\ell+1)\dots\tau\rho(n) \right\rangle \\ &= \left\langle [B(\tilde{1}) + i\beta\omega_1\rho(\tilde{1})][B(\tilde{2}) + i\beta\omega_2\rho(\tilde{2})] \right. \\ &\quad \left. \dots [B(\tilde{\ell}) + i\beta\omega_\ell\rho(\tilde{\ell})]\rho(\ell+1)\dots\rho(\tilde{n}) \right\rangle \end{aligned} \quad (3.17)$$

where $\tilde{j} = q_j, -\omega_j$. Multiplying these out, each correlation function has arguments with tildes. Using the result

$$\tau_1\tau_2\mathcal{A}^* = \mathcal{A} \quad (3.18)$$

and

$$\tau_1\tau_2 B^*(1) = B(\tilde{1}) \quad (3.19)$$

$$\tau_1\tau_2 \rho^*(1) = \rho(\tilde{1}) \quad (3.20)$$

we find the simple result

$$\begin{aligned} C_{BB\dots B\rho\dots\rho}(12\dots\ell\ell+1\dots n) &= \left[\left\langle [B(1) - i\beta\omega_1\rho(1)][B(2) - i\beta\omega_2\rho(2)] \right. \right. \\ &\quad \left. \left. \dots [B(\ell) - i\beta\omega_\ell\rho(\ell)]\dots\rho(\ell+1)\dots\rho(n) \right\rangle \right]^* \quad . \end{aligned} \quad (3.21)$$

If we define

$$\psi(i) = B(i) - i\beta\omega_i\rho(i) \quad , \quad (3.22)$$

the mixed correlation function is obtained as

$$C_{BB\dots B\rho\rho}(12\dots\ell, \ell+1\dots n) = \left\langle \psi(1)\psi(2)\dots\psi(\ell)\rho(\ell+1)\dots\rho(n) \right\rangle \quad (3.23)$$

One easily sees that this holds for the cumulants[8]

$$G_{BB\dots B\rho\rho}(12\dots\ell, \ell+1\dots n) = G_{\psi\psi\dots\psi\rho\dots\rho}^*(12\dots\ell, \ell+1\dots n) \quad (3.24)$$

and for the single-particle quantities

$$\mathcal{G}_{BB\dots B\rho\rho}(12\dots\ell, \ell+1\dots n) = \tilde{\mathcal{G}}_{\psi\psi\dots\psi\rho\dots\rho}^*(12\dots\ell, \ell+1\dots n) \quad . \quad (3.25)$$

1. Two-point cumulants

Using the FD relations at the two-point level we find

$$G_{B\rho}(12) = G_{B\rho}^*(12) + i\beta\omega_1 G_{\rho\rho}^*(12) \quad . \quad (3.26)$$

Due to translational invariance in space and time

$$G_{\alpha\beta}(12) = G_{\alpha\beta}(1)\delta(1+2) \quad (3.27)$$

$$G_{\rho B}(1) = G_{B\rho}^*(1) \quad . \quad (3.28)$$

We see that Eq.(3.26) can be written as

$$G_{B\rho}(1) = G_{\rho B}(1) + i\beta\omega_1 G_{\rho\rho}^*(1) \quad . \quad (3.29)$$

Since $G_{\rho\rho}(1)$ is real, using eqns. (3.28) and (3.29), we have the conventional result

$$Im G_{B\rho}(1) = -Im G_{\rho B}(1) = \frac{\beta\omega_1}{2} G_{\rho\rho}(1) \quad (3.30)$$

which can be used to construct the dispersion relation

$$G_{\rho B}(1) = \int \frac{d\bar{\omega}}{2\pi} \frac{\beta \bar{\omega} G_{\rho\rho}(q_1, \bar{\omega})}{\omega_1 - \bar{\omega} + i\eta} \quad . \quad (3.31)$$

Taking the inverse time Fourier transform we find

$$G_{\rho B}(q, t) = \theta(t) \beta \frac{\partial}{\partial t} G_{\rho\rho}(q, t) \quad . \quad (3.32)$$

Using the FD relation (3.31) we obtain for $G_{\rho B}(q, 0)$

$$G_{\rho B}(q, \omega = 0) = \int \frac{d\bar{\omega}}{2\pi} \frac{\beta \bar{\omega} G_{\rho\rho}(q_1, \bar{\omega})}{-\bar{\omega} + i\eta} = -\beta \bar{\rho} S(q) \quad (3.33)$$

where $S(q)$ is the static structure factor at wave vector q and $\bar{\rho}$ is the average density so that the RHS represents the correlation of density fluctuations at equal time. From eqn. (3.29), we obtain the very useful result

$$i\beta\omega G_{\rho\rho}(q, \omega) + G_{\rho B}(q, \omega) - G_{B\rho}(q, \omega) = 0 \quad (3.34)$$

Eqn. (3.34) is written in a compact form as $\mathcal{F}[G] = 0$, where the functional \mathcal{F} acting on the elements of a 2×2 matrix $A_{\mu\nu}$ is defined as follows

$$\mathcal{F}[A] \equiv \text{Tr} [(i\beta\omega \mathcal{I} + \varepsilon) \cdot A(q, \omega)] \quad . \quad (3.35)$$

We have introduced the traceless anti-symmetric matrix ε in the RHS of eqn. (fdr-func) :

$$\varepsilon_{B\rho} = 1, \quad \varepsilon_{\rho B} = -1, \quad \varepsilon_{\mu\mu} = 0. \quad (3.36)$$

\mathcal{I} is the 2×2 identity matrix. The functional $\tilde{\mathcal{F}}$ is defined as

$$\tilde{\mathcal{F}}[A] \equiv \text{Tr} [(-i\beta\omega \mathcal{I} + \tilde{\varepsilon}) \cdot A(q, \omega)] \quad (3.37)$$

where the transpose matrix $\tilde{\varepsilon}_{\mu\nu} = \varepsilon_{\nu\mu}$. Vanishing of the above defined functional $\mathcal{F}[A]$ then ensures the fluctuation-dissipation relation (FDR) among the elements of the matrix A . It is straightforward to show that the relation

$$\mathcal{F}[A] = 0 \quad (3.38)$$

is satisfied for the two-point propagator A belonging to any member of the set $\{G^{(0)}, G, \mathcal{G}\}$. With the condition (3.38) the elements of A will be referred to as FDR matrix propagator

(FDRMP). We show in Appendix C how to build new propagators which also obey the same FDR. If $A_{\alpha\beta}(q, \omega)$ and $D_{\alpha\beta}(q, \omega)$ are respectively two FDRMP then the matrix $H_{\alpha\beta}$ defined in terms of the interaction matrix $\sigma_{\mu\nu}$ as

$$H_{\alpha\beta}(q, \omega) = \sum_{\delta\gamma} A_{\alpha\delta}(q, \omega) \sigma_{\delta\gamma}(q) D_{\gamma\beta}(q, \omega) \quad (3.39)$$

is also a FDRMP since it satisfies the relation $\mathcal{F}[H_{\alpha\beta}] = 0$. This property of composite propagators following the same Fluctuation-dissipation relation is an important ingredient in the analysis of memory functions discussed later in this work.

2. Two-point Vertices

The two-point vertices $\Gamma_{\alpha\beta}(1)$ are defined as the corresponding elements of the inverse of the general two-point correlation matrix $G_{\alpha\beta}$:

$$\sum_{\mu} \Gamma_{\alpha\mu}(1) G_{\mu\beta}(1) = \delta_{\alpha\beta} \quad . \quad (3.40)$$

For $\alpha = \rho$ and $\beta = B$, using $G_{BB}(1) = 0$ [17], it follows from the above equation that $\Gamma_{\rho\rho}(1) = 0$. For other choices of α, β we obtain,

$$\Gamma_{\rho B}(1) G_{B\rho}(1) = 1 \quad (3.41)$$

$$\Gamma_{B\rho}(1) G_{\rho B}(1) = 1 \quad (3.42)$$

$$\Gamma_{BB}(1) G_{B\rho}(1) + \Gamma_{B\rho}(1) G_{\rho\rho}(1) = 0 \quad . \quad (3.43)$$

Using Eqs. (3.41) and (3.42), Eq. (3.43) can be written in the form

$$G_{\rho\rho}(1) = -G_{\rho B}(1) \Gamma_{BB}(1) G_{B\rho}(1) \quad . \quad (3.44)$$

We easily find

$$\Gamma_{B\rho}(q, \omega) = \Gamma_{\rho B}^*(q, \omega) \quad (3.45)$$

Starting with Eq.(3.26):

$$G_{B\rho}(1) = G_{B\rho}^*(1) + i\beta\omega_1 G_{\rho\rho}^*(1) \quad (3.46)$$

we can use Eq.(3.41), (3.42) and (3.45) to write

$$\frac{1}{\Gamma_{\rho B}(1)} = \frac{1}{\Gamma_{B\rho}(1)} + i\beta\omega_1 G_{\rho\rho}(1) \quad (3.47)$$

Furthermore using Eq.(3.43) and Eq.(3.47), and canceling a common denominator, gives

$$i\beta\omega_1 \Gamma_{BB}(1) = \Gamma_{\rho B}(1) - \Gamma_{B\rho}(1) \quad . \quad (3.48)$$

In terms of the functional \mathcal{F} defined in Eq. (3.35) the above relation is obtained as

$$\mathcal{F}[\Gamma] = 0 \quad , \quad (3.49)$$

where the matrix vertex $\Gamma_{\alpha\beta}$ is defined in the $\{\rho, B\}$ domain.

3. FD symmetry and Three-point cumulants

There are substantial differences in specific cumulants for the different realizations of simple-fluid dynamics. For example the noninteracting three-point cumulant $G_{B\rho\rho}^{(0)}(123)$ for SD and ND are quite different. Despite this fact we find that both sets of cumulants satisfy the same fluctuation dissipation relations.

In treating the three-point cumulants involving the fields at three different points, we adopt the following notation (in Fourier space)

$$G_{\rho_i \rho_j \rho_k} \equiv G_{\rho\rho\rho}(ijk) = G_{\rho\rho\rho}(jki) = G_{\rho\rho\rho}(kji) \quad (3.50)$$

$$G_{\rho_i \rho_j B_k} \equiv G_{\rho\rho B}(ijk) = G_{\rho B\rho}(jki) = G_{B\rho\rho}(kij), \quad (3.51)$$

$$G_{B_i B_j B_k} \equiv G_{BBB}(ijk) = G_{BBB}(jki) \text{ etc.} \quad (3.52)$$

where the set $\{ijk\} \implies \{123\}, \{231\},$ and $\{321\}$. The factor β will for now be absorbed in the frequency ω . For cumulants involving three B fields we obtain using the basic fluctuation-dissipation relation (3.26) the following relations :

$$\begin{aligned}
G_{B_i B_j B_k} &= G_{B_i B_j B_k}^* + i\omega_i G_{\rho_i B_j B_k}^* + i\omega_j G_{B_i \rho_j B_k}^* + i\omega_k G_{B_i B_j \rho_k}^* \\
&\quad - \omega_i \omega_j G_{\rho_i \rho_j B_k}^* - \omega_j \omega_k G_{B_i \rho_j \rho_k}^* - \omega_k \omega_i G_{\rho_k B_j B_i}^* - i\omega_i \omega_j \omega_k G_{\rho_i \rho_j \rho_k}^*
\end{aligned} \tag{3.53}$$

Since $G_{BBB} = G_{BBB}^* = 0$, using this in the complex conjugate of Eqn. (3.53) gives the relation

$$\begin{aligned}
0 &= i\omega_i G_{\rho_i B_j B_k} + i\omega_j G_{B_i \rho_j B_k} + i\omega_k G_{B_i B_j \rho_k} + \omega_i \omega_j G_{\rho_i \rho_j B_k} \\
&\quad + \omega_j \omega_k G_{B_i \rho_j \rho_k} + \omega_k \omega_i G_{\rho_k B_j \rho_i} - i\omega_i \omega_j \omega_k G_{\rho_i \rho_j \rho_k}
\end{aligned} \tag{3.54}$$

Similarly for the cumulants respectively involving two and one B fields are obtained as follows :

$$G_{B_i B_j \rho_k} = G_{B_i B_j \rho_k}^* + i\omega_i G_{\rho_i B_j \rho_k}^* + i\omega_j G_{B_i \rho_j \rho_k}^* - \omega_i \omega_j G_{\rho_i \rho_j \rho_k}^* \tag{3.55}$$

$$G_{B_i \rho_j \rho_k} = G_{B_i \rho_j \rho_k}^* + i\omega_i G_{\rho_i \rho_j \rho_k}^* . \tag{3.56}$$

To simplify the above relations involving the three-point cumulants, we define $\mu_i = i\omega_i G_{\rho_i B_j B_k}$, $\sigma_i = \omega_j \omega_k G_{B_i \rho_j \rho_k}$, and $\nu = \omega_i \omega_j \omega_k G_{\rho_i \rho_j \rho_k}$. In terms of the newly defined quantities we rewrite the Eqs. (3.55)-(3.54) respectively as

$$-\mu_k^* = \mu_k + \sigma_j + \sigma_i - i\nu \tag{3.57}$$

$$\sigma_i = \sigma_i^* + i\nu \tag{3.58}$$

$$\mu_i + \mu_j + \mu_k + \sigma_i + \sigma_j + \sigma_k = i\nu \tag{3.59}$$

where ν is real. Solving the above equations we obtain

$$\sigma_i'' = \nu/2 \tag{3.60}$$

$$\mu_i' = -\frac{\sigma_j' + \sigma_k'}{2} \tag{3.61}$$

$$\mu_T'' = \frac{\nu}{2} \tag{3.62}$$

where $\mu_T = \mu_i + \mu_j + \mu_k$ is the sum of the μ 's. These results seem rather general.

4. FD symmetry and Three-point Cumulants

Next we analyze the relations between the different three-point vertex functions. For this we use the general definition of the three-point cumulants and the corresponding three-point vertex function as,

$$G_{\alpha_i \mu_j \nu_k} = -G_{\alpha \alpha'}(i) G_{\mu \mu'}(j) G_{\nu \nu'}(k) \Gamma_{\alpha'_i \mu'_j \nu'_k} \quad (3.63)$$

where α_i stands for the field α at the point i and so on. The fields α, μ and ν are respectively taken from the set $\{\rho, B\}$. In the RHS of Eq. (3.63) we have used the notation $G_{\mu\nu}(i)$ following Eq. (3.26), *i.e.*, $G_{\mu\nu}(ij) = G_{\mu\nu}(i)\delta(i+j)$. Since $G_{BB} = 0$, taking all the three fields to be B at the respective points i, j , and k we obtain, ,

$$G_{B_i B_j B_k} = -G_{B\rho}(i) G_{B\rho}(j) G_{B\rho}(k) \Gamma_{\rho_i \rho_j \rho_k} \quad (3.64)$$

Since $G_{B_i B_j B_k} = 0$ it follows from the last equation that $\Gamma_{\rho_i \rho_j \rho_k} = 0$. The expression (3.63) for the various three-point cumulants in terms of two-point cumulants simplifies due to the vanishing of $\Gamma_{\rho\rho\rho}$. Let us first consider the expression (3.54) involving *three* B fields. In the appendix A we show that by a) using the expression (3.63) for $G_{\alpha\mu\nu}$ in Eq. (3.54), and b) by collecting the various coefficients of the different vertices Γ 's, we obtain the following relation between the three-point vertices.

$$\begin{aligned} & i \left(\omega_i \Gamma_{B_i \rho_j \rho_k} + \omega_j \Gamma_{\rho_i B_j \rho_k} + \omega_k \Gamma_{\rho_i \rho_j B_k} \right) - i \omega_i \omega_j \omega_k \Gamma_{B_i B_j B_k} \\ & + \left\{ \omega_i \omega_j \Gamma_{B_i B_j \rho_k} + \omega_j \omega_k \Gamma_{\rho_i B_j B_k} + \omega_k \omega_i \Gamma_{B_i \rho_j B_k} \right\} = 0 \quad . \end{aligned} \quad (3.65)$$

Next, we consider the relations (3.55) which contain three-point cumulants involving correlation of *two* B fields.

$$G_{B_i B_j \rho_k}^* = G_{B_i B_j \rho_k} - i \omega_i G_{\rho_i B_j \rho_k} - i \omega_j G_{B_i \rho_j \rho_k} - i \omega_i \omega_j G_{\rho_i \rho_j \rho_k} \quad (3.66)$$

Using the the three-point cumulants corresponding to this case in terms of vertex functions as defined in Eq. (3.63) and collecting the coefficients of the various vertex functions, we obtain in Appendix A the result :

$$i \omega_k \Gamma_{\rho_i \rho_j B_k}^* + i \omega_i \Gamma_{B_i \rho_j \rho_k} + i \omega_j \Gamma_{\rho_i B_j \rho_k} + \omega_i \omega_j \Gamma_{B_i B_j \rho_k} = 0 \quad . \quad (3.67)$$

By doing circular permutations of the indices $\{i, j, k\}$ in Eq. (3.67) we obtain the following relations involving the vertices with one B -field leg:

$$i\omega_i \Gamma_{B_i \rho_j \rho_k}^* + i\omega_j \Gamma_{\rho_i B_j \rho_k} + i\omega_k \Gamma_{\rho_i \rho_j B_k} + \omega_j \omega_k \Gamma_{\rho_i B_j B_k} = 0 \quad (3.68)$$

$$i\omega_j \Gamma_{\rho_i B_j \rho_k}^* + i\omega_k \Gamma_{\rho_i \rho_j B_k} + i\omega_i \Gamma_{B_i \rho_j \rho_k} + \omega_k \omega_i \Gamma_{B_i \rho_j B_k} = 0 \quad (3.69)$$

The above relations are further simplified with the following notations :

$$\phi_i = i\omega_i \Gamma_{B_i \rho_j \rho_k}, \quad (3.70)$$

$$\psi_i = \omega_j \omega_k \Gamma_{\rho_i B_j B_k}, \quad (3.71)$$

$$P = -i\omega_i \omega_j \omega_k \Gamma_{B_i B_j B_k} . \quad (3.72)$$

We obtain from Eq. (3.65) using the above notation the result

$$P + \phi_T + \psi_T = 0 \quad (3.73)$$

where $\phi_T = \phi_1 + \phi_2 + \phi_3$ and $\psi_T = \psi_1 + \psi_2 + \psi_3$. In the simplified notation the Eqs. (3.67) - (3.69) respectively reduces to the cyclic forms

$$-\phi_i^* + \phi_j + \phi_k + \psi_i = 0 . \quad (3.74)$$

Looking at the imaginary parts of these equations we have $\psi_i'' = -\phi_T''$, while for the real parts $-\phi_i' + \phi_j' + \phi_k' + \psi_i' = 0$. The last equation has the simple solution

$$\phi_j' = -\frac{1}{2}(\psi_i' + \psi_k') \quad (3.75)$$

In terms of the original notation, using the defining relation (3.63) for the different vertices, we obtain the following set of *nonperturbative* relations among the various vertex functions.

$$\begin{aligned} \Gamma_{B_i \rho_j \rho_k}'' &= \frac{1}{2}(\omega_k \Gamma_{B_i \rho_j B_k}' + \omega_j \Gamma_{B_i B_j \rho_k}') \\ \omega_j \omega_k \Gamma_{\rho_i B_j B_k}'' &= \omega_i \omega_k \Gamma_{B_i \rho_j B_k}'' = \omega_i \omega_j \Gamma_{B_i B_j \rho_k}'' \\ &= -\left(\omega_i \Gamma_{B_i \rho_j \rho_k}' + \omega_j \Gamma_{\rho_i B_j \rho_k}' + \omega_k \Gamma_{\rho_i \rho_j B_k}' \right) . \end{aligned} \quad (3.76)$$

The vertex function $\Gamma_{B_i B_j B_k}$ is real, *i.e.*, $\Gamma''_{B_i B_j B_k} = 0$. The real part satisfies the relation

$$\omega_i \omega_j \omega_k \Gamma'_{B_i B_j B_k} = -2 \left[\omega_i \Gamma'_{B_i \rho_j \rho_k} + \omega_j \Gamma'_{\rho_i B_j \rho_k} + \omega_k \Gamma'_{\rho_i \rho_j B_k} \right] . \quad (3.77)$$

IV. KINETIC EQUATIONS

We now present starting from the basic equation (3.40), the derivation of a kinetic equation for time evolution of the density autocorrelation function $G_{\rho\rho}(q, t)$. This kinetic equation involves the different contributions to the vertex functions Γ_{ij} . The analysis is similar to the discussions in SDENE[6] and MMS[7]. These developments start with a discussion of the form of $\Gamma_{\alpha\beta}(q, \omega)$ in the time domain. If we look at

$$\Gamma_{B\rho}(q, \omega) = \frac{1}{G_{\rho B}(q, \omega)} . \quad (4.1)$$

The response function $G_{\rho B}$ vanish algebraically as $\omega \rightarrow \infty$, and hence the two-point vertex $\Gamma_{B\rho}$ diverges in this limit. We assume that

$$\lim_{\omega \rightarrow \infty} \Gamma_{B\rho}(q, \omega) = -D_q \omega^2 + i\omega A_q + C_q \quad (4.2)$$

where the coefficients A , C and D are real and not universal. We define the "local" quantity

$$\Gamma_{B\rho}^{(\ell)}(q, \omega) = -D_q \omega^2 + i\omega A_q + C_q \quad (4.3)$$

for all frequencies. The corresponding subtracted quantities are obtained as

$$\Gamma_{B\rho}^{(s)}(q, \omega) = \Gamma_{B\rho}(q, \omega) - \Gamma_{B\rho}^{(\ell)}(q, \omega) \quad (4.4)$$

which vanish for large frequencies. At low frequencies

$$\Gamma_{B\rho}^{(s)}(q, \omega = 0) = \Gamma_{B\rho}(q, \omega = 0) - \Gamma_{B\rho}^{(\ell)}(q, \omega = 0) . \quad (4.5)$$

Using the Eqs. (3.33) and (4.1), we obtain $\Gamma_{B\rho}(q, 0) = -[\bar{\rho}\beta S(q)]^{-1}$. From Eq. (4.3) it follows that $\Gamma_{B\rho}^{(\ell)}(q, 0) = C_q$ and hence using Eq. (4.4) we obtain

$$\Gamma_{B\rho}^{(s)}(q, \omega = 0) = -C_q - [\bar{\rho}\beta S(q)]^{-1} . \quad (4.6)$$

Next, assuming that the FDR holds locally, we obtain from Eq. (4.3) and FD relation (3.48)

$$\omega \Gamma_{BB}^{(\ell)}(q, \omega) = -2\omega A_q \quad (4.7)$$

Hence $\Gamma_{BB}^{(\ell)}(q, 0) = -2\beta^{-1}A_q$. Assuming $A_q \geq 0$ and taking inverse Fourier transforms we obtain in the time space the following results.

$$\hat{\Gamma}_{BB}^{(\ell)}(q, t - t') = -\beta^{-1}A_q \delta(t - t') \quad (4.8)$$

$$\hat{\Gamma}_{B\rho}^{(\ell)}(q, t - t') = \left[D_q \frac{\partial^2}{\partial t^2} - A_q \frac{\partial}{\partial t} + C_q \right] \delta(t - t') \quad (4.9)$$

where the Γ 's in the time space are denoted with a hat. For the full two-point vertex functions $\Gamma_{B\rho}$ and Γ_{BB} the following relations involving the local and subtracted parts,

$$\hat{\Gamma}_{B\rho}(q, t - t') = \hat{\Gamma}_{B\rho}^{(\ell)}(q, t - t') + \hat{\Gamma}_{B\rho}^{(s)}(q, t - t') \quad (4.10)$$

$$\hat{\Gamma}_{BB}(q, t - t') = -\beta^{-1}A_q \delta(t - t') + \hat{\Gamma}_{BB}^{(s)}(q, t - t') \quad (4.11)$$

It then follows for the subtracted parts $\Gamma_{B\rho}^{(s)}$ that we have the dispersion relation (since the FDR holds locally for $\Gamma_{B\rho}^{(\ell)}$)

$$\Gamma_{B\rho}^{(s)}(q, \omega) = \int \frac{d\bar{\omega}}{2\pi} \frac{\beta \bar{\omega} \Gamma_{BB}^{(s)}(q, \bar{\omega})}{\omega - \bar{\omega} + i\eta} \quad (4.12)$$

reflecting the fact that the vertex function $\Gamma_{B\rho}$ is analytic in the upper half plane. From the above FDR it follows directly that

$$\Gamma_{B\rho}^{(s)}(q, \omega = 0) = \beta \hat{\Gamma}_{BB}^{(s)}(q, 0) \quad (4.13)$$

In the time domain the FDR (4.12) reads

$$\hat{\Gamma}_{B\rho}^{(s)}(q, t - t') = \beta \theta(t - t') \frac{\partial}{\partial t} \hat{\Gamma}_{BB}^{(s)}(q, t - t') \quad (4.14)$$

A. Memory Function Equation

The density auto correlation function $G_{\rho\rho}$ is of particular interest in the present theoretical model for studying the slow dynamics of a dense liquid. The natural order parameter for

the ENE transition which is the primary focus of this paper is the long-time limit of this function. In this section we obtain an equation of motion for the density correlation function. First we express Eq.(3.43) in the time-domain

$$\hat{\Gamma}_{B\rho}^{(\ell)}(q, 0)G_{\rho\rho}(q, t - t') + \hat{\Gamma}_{BB}^{(\ell)}(q, 0)G_{B\rho}(q, t - t') = \Psi_q(t, t') \quad (4.15)$$

where

$$\Psi(t, t') = - \int_{-\infty}^t ds \hat{\Gamma}_{B\rho}^{(s)}(t - s)G_{\rho\rho}(s - t') - \int_{-\infty}^{t'} ds \hat{\Gamma}_{BB}^{(s)}(t - s)G_{\rho B}(t' - s) \quad . \quad (4.16)$$

In writing Eq. (4.15) we have used the fact that the response functions are time ordered so that $\hat{\Gamma}_{B\rho}^{(s)}(t - s) \sim \theta(t - s)$ and $G_{\rho B}(t' - s) \sim \theta(t' - s)$. We then use the fluctuation dissipation relations (3.32) and (4.14) to obtain

$$-\Psi(t, t') = \int_{-\infty}^t ds \left[\frac{\partial}{\partial s} \beta \hat{\Gamma}_{BB}^{(s)}(t - s) \right] G_{\rho\rho}(s - t') + \int_{-\infty}^{t'} ds \hat{\Gamma}_{BB}^{(s)}(t - s) \frac{\partial}{\partial s} \beta G_{\rho\rho}(t' - s) \quad . \quad (4.17)$$

Integrating by parts[18] in the first integral gives

$$\begin{aligned} -\Psi(t, t') &= \beta \hat{\Gamma}_{BB}^{(s)}(0)G_{\rho\rho}(t - t') - \int_{-\infty}^t ds \beta \hat{\Gamma}_{BB}^{(s)}(t - s) \frac{\partial}{\partial s} G_{\rho\rho}(s - t') \\ &\quad + \int_{-\infty}^{t'} ds \beta \hat{\Gamma}_{BB}^{(s)}(t - s) \frac{\partial}{\partial s} G_{\rho\rho}(t' - s) \\ &= \beta \hat{\Gamma}_{BB}^{(s)}(0)G_{\rho\rho}(t - t') - \int_{t'}^t ds \beta \hat{\Gamma}_{BB}^{(s)}(t - s) \frac{\partial}{\partial s} G_{\rho\rho}(t' - s) \end{aligned} \quad (4.18)$$

where we have assumed that $t > t'$. We then have the kinetic equation

$$\left[\hat{\Gamma}_{B\rho}^{(\ell)}(q, 0) + \beta \hat{\Gamma}_{BB}^{(s)}(q, 0) \right] G_{\rho\rho}(q, t - t') - \int_{t'}^t ds \beta \hat{\Gamma}_{BB}^{(s)}(q, t - s) \frac{\partial}{\partial s} G_{\rho\rho}(q, t' - s) = 0 \quad . \quad (4.19)$$

Since $t > t'$ we have dropped the $\hat{\Gamma}_{BB}^{(\ell)}(q, 0)G_{B\rho}(q, t - t')$ term in the LHS of Eq. (4.15). Using the relations (4.13) and (4.6) we obtain for the quantity within square brackets on the LHS of (4.19) as

$$\hat{\Gamma}_{B\rho}^{(\ell)}(q, 0) + \beta \hat{\Gamma}_{BB}^{(s)}(q, 0) = \hat{\Gamma}_{B\rho}^{(\ell)}(q, 0) + \Gamma_{B\rho}^{(s)}(q, \omega = 0) \quad . \quad (4.20)$$

Using the relations (4.6) and (4.9) respectively for the second and the first terms on the RHS, the kinetic equation then takes the form

$$\left[D_q \frac{\partial^2}{\partial t^2} - A_q \frac{\partial}{\partial t} - \frac{1}{\bar{\rho} \beta S(q)} \right] G_{\rho\rho}(q, t) + \int_{t'}^t ds \beta \hat{\Gamma}_{BB}^{(s)}(q, t-s) \frac{\partial}{\partial s} G_{\rho\rho}(q, t'-s) = 0 \quad . \quad (4.21)$$

We see that our dynamical problem is now in the form of a memory function equation and the dynamic part of the memory function is given by the subtracted part $\hat{\Gamma}_{BB}^{(s)}$ of the vertex function. The static structure factor is the same for all fluids with the same pair potential. The coefficients A_q and D_q can be constructed using perturbation theory. In the case of ND we have the ideal gas result:

$$\Gamma_{B\rho}^{(0)}(q, \omega) = -\frac{1}{\beta \bar{\rho} \mathcal{S}^*(z)} \quad (4.22)$$

where $z = \omega/(\sqrt{2}qv_0)$, $v_0 = \sqrt{k_B T/m}$ is the thermal velocity and

$$\mathcal{S}(x) = 1 - 2xe^{-x^2} \int_0^x du e^{u^2} - i\sqrt{\pi}xe^{-x^2} \quad . \quad (4.23)$$

In the large frequency limit

$$\Gamma_{B\rho}^{(0)}(q, \omega) = -\frac{1}{\beta \bar{\rho}} \left[-\left(\frac{\omega}{qv_0} \right)^2 + 1 \right] \quad (4.24)$$

and on comparing this relation with Eq. (4.3) we can identify at the lowest order,

$$A_0(q) = 0, \quad (4.25)$$

$$C_0(q) = -\frac{1}{\beta \bar{\rho}}, \quad (4.26)$$

$$q^2 D_0(q) = \beta m C_0(q) = -\frac{m}{\bar{\rho}} \quad (4.27)$$

where the equilibrium density is given by $\bar{\rho}$. With the above identification we obtain the kinetic Eq. (4.21) in the form

$$\left[\frac{\partial^2}{\partial t^2} + \Omega_q^2 \right] G_{\rho\rho}(q, t) + \int_0^t ds \hat{\Gamma}_{\text{mc}}(q, t-s) \frac{\partial}{\partial s} G_{\rho\rho}(q, t'-s) = 0 \quad . \quad (4.28)$$

where we have defined

$$\Omega_q^2 = \frac{q^2}{\beta m S(q)} \equiv q^2 c^2(q) \quad (4.29)$$

$$\Gamma_{\text{mc}}(q, t) = -\frac{\beta \bar{\rho}}{m} q^2 \hat{\Gamma}_{BB}^{(s)}(q, t). \quad (4.30)$$

Note that this resembles the second-order oscillator equation of standard MCT [10, 11] *without* the bare dissipation coefficient. In the case of Newtonian dynamics the microscopic dynamics is reversible and the kinetic equation (4.28) has been obtained without projection in to a space of slow modes. Indeed as we discuss below in order for the ENE transition to take place we need to break the time-reversal symmetry.

The kinetic equation is diagonalized using a Laplace transform defined as

$$\tilde{G}_{\rho\rho}(q, z) = -i \int_0^\infty dt e^{izt} G_{\rho\rho}(q, t) \quad . \quad (4.31)$$

The Laplace transformed kinetic equation (4.21) reduces to the form for the normalized density correlation function $F(q, t)$

$$F(q, z) = \frac{\tilde{G}_{\rho\rho}(q, z)}{\bar{\rho} S(q)} = \frac{z + iq^2 \Gamma_{\text{mc}}(q, z)}{z[z + iq^2 \Gamma_{\text{mc}}(q, z)] - \Omega_q^2} \quad (4.32)$$

where $\bar{\rho} S(q)$ in the denominator of the LHS above is the equal-time correlation function and normalize $F(q, t)$ to unity. We obtain the following integral equation for $F(q, z)$

$$\begin{aligned} \frac{F(q, z)}{1 - zF(q, z)} &= -\frac{iq^2}{\Omega_q^2} \int_0^\infty dt e^{izt} \beta \Gamma_{\text{mc}}(q, t) \\ &= i\bar{\rho} S(q) \int_0^\infty dt e^{izt} \beta^2 \Gamma_{BB}^{(s)}(q, t) \\ &= -\bar{\rho} S(q) \beta^2 \Gamma_{BB}^{(s)}(q, z) \quad . \end{aligned} \quad (4.33)$$

We now turn to the mechanism which produces large $F(q, z)$ and $\Gamma_{BB}^{(s)}(q, z)$ as $z \rightarrow 0$. This involves determining the memory function as a functional of $F(q, t)$. We define $F(q)$ as the long time limit of the normalized density correlation function $F(q, t)$ to denote the so called non ergodicity parameter.

$$\lim_{z \rightarrow 0} \{zF(q, z)\} \rightarrow F(q) \quad (4.34)$$

From Eq. (4.33) we obtain the following integral equation for $F(q)$

$$\frac{F(q)}{1-F(q)} = \bar{\rho}S(q)\beta^2\Gamma_{BB}^{(s)}(q) \quad . \quad (4.35)$$

$\Gamma_{BB}^{(s)}(q)$ is a functional of $F(q)$

$$\lim_{z \rightarrow 0} \{-z\Gamma_{BB}^{(s)}(q, z)\} \rightarrow \Gamma_{BB}^{(s)}(q) \quad (4.36)$$

and is obtained from the explicit evaluation of the subtracted part of the vertex function Γ_{BB} in a perturbation series. Solution of the resulting self-consistent equation, Eq.(4.35), for $F(q)$ corresponds to what we call the ENE problem. This now primarily involves expressing $\Gamma_{BB}^{(s)}(q, t)$ self consistently in terms of the correlation functions. The self-consistent treatment of the dynamics constitutes the basic feedback mechanism and gives rise to the slow dynamics characteristics of the mode coupling theories[10, 11].

V. SELF-CONSISTENT DEVELOPMENT

A. General

In Ref. 1 we obtained that the canonical partition function for the N particle classical system following Newtonian dynamics (ND) in the form

$$Z_N[H, h, \hat{h}] = \int \prod_{i=1}^N \mathcal{D}(\Psi_i) \mathcal{D}(\hat{\Psi}_i) \mathcal{D}(\Psi_i^{(0)}) e^{-\mathcal{A}_0 - \mathcal{A}_I + H \cdot \phi + h \cdot \psi + \hat{h} \cdot \hat{\psi}} \quad (5.1)$$

where $\{\Psi_i, \hat{\Psi}_i\}$ are respectively the microscopic variables for the i -th particle ($i = 1, \dots, N$) and its hatted counterpart in a Martin Siggia Rose (MSR) field theoretic formulation of the problem. In the above expression for Z_N , the superscript 0 in $\Psi_i^{(0)}$ in the RHS refers to the initial state of the system. The MSR action is written as a sum of two parts as stated in eqn. (2.14). \mathcal{A}_0 is the quadratic part of the action including the quadratic contribution to the initial probability distribution. \mathcal{A}_I is the interaction part of the MSR action given in eqn. (2.15). The set $\{h_i, \hat{h}_i\}$ denote the currents conjugate to $\{\Psi_i, \hat{\Psi}_i\}$ for $i = 1, \dots, N$ respectively. The collective field Φ_α is defined in eqn. (2.10) and H_α is current conjugate to Φ_α . The canonical partition function (5.1) can be written in the convenient form

$$Z_N = \text{Tr}^{(N)} e^{-A_I + H \cdot \Phi} \quad (5.2)$$

where we have introduced the N particle average as,

$$\text{Tr}^{(N)} = \int \prod_{i=1}^N \mathcal{D}(\Psi_i) \mathcal{D}(\hat{\Psi}_i) d\Psi_i^{(0)} e^{-A_0} . \quad (5.3)$$

The partition for the interacting system is obtained using the identity

$$e^{-A_I + H \cdot \Phi} = e^{\hat{A}_T} e^{H \cdot \Phi} \quad (5.4)$$

in terms of operator

$$\hat{A}_T = \frac{1}{2} \int d1 \int d2 \sum_{\alpha, \beta} \hat{H}_\alpha(1) \sigma_{\alpha\beta}(12) \hat{H}_\beta(2) \quad (5.5)$$

where $\hat{H}_\alpha(1) = [\delta/\delta\hat{H}_\alpha(1)]$. We rewrite the partition function in a form that allows us to formally carry out the average in eqn. (5.2) using the functional identity (5.4).

$$Z_N = e^{\hat{A}_T} \text{Tr}^{(N)} e^{H \cdot \Phi} . \quad (5.6)$$

The sum over the degrees of freedom in eqn. (2.11) factorizes into a product of sums over the degrees of freedom of each particle. Together, these observations lead to the following result for the noninteracting Partition function,

$$Z_N^{(0)} = \text{Tr}^{(N)} e^{H \cdot \Phi} = (Z_1)^N . \quad (5.7)$$

where Z_1 is the single particle partition function.

$$Z_1 = \text{Tr}^{(1)} e^{H \cdot \Phi} \quad (5.8)$$

Working in the grand canonical ensemble, the grand partition function for the interacting problem given by (2.5) is obtained in the form

$$Z_T[H, h, \hat{h}] = \sum_{N=0}^{\infty} \frac{\rho_0^N}{N!} Z_N[H, h, \hat{h}] = e^{\hat{A}_T} e^{W_0} , \quad (5.9)$$

in terms of the single particle function W_0 . In eqn. (5.9), $z = \rho_0$ is the fugacity. The cumulants of the fields Φ_α are generated by taking functional derivatives of the generating functional $W[H] = \ln Z_T$ with respect the fields H introduced in eqn. (2.19) above.

It was shown in FTSPD that the one-point average G_α in a field Φ_α satisfies the fundamental identity

$$G_\alpha = \text{Tr} \phi_\alpha e^{H \cdot \phi + \Delta W[H]}. \quad (5.10)$$

where Tr is the sum over all the degrees of freedom including the conjugate MSR degrees of freedom for a *single* particle. The functional ΔW is defined by

$$\Delta W[H] = W[H + F] - W[H] \quad (5.11)$$

where

$$F_\alpha = \sum_\beta \sigma_{\alpha\beta} \phi_\beta \quad . \quad (5.12)$$

The dependence of the theory on the interaction potential is controlled by $\Delta W[H]$. This quantity is expressed in a functional Taylor-series expansion in powers of the potential V as,

$$\Delta W[H] = \sum_\alpha F_\alpha \frac{\delta}{\delta H_\alpha} W[H] + \sum_{\alpha\beta} \frac{1}{2} F_\alpha F_\beta \frac{\delta^2}{\delta H_\alpha \delta H_\beta} W[H] + \dots \quad (5.13)$$

with F_α given by Eq.(5.12). For systems fluctuating in equilibrium the force matrix is given in terms of fourier transforms by eqn. (2.16) above,

$$\sigma_{\alpha\beta}(q) = V(q) [\delta_{\alpha\rho} \delta_{\beta B} + \delta_{\alpha B} \delta_{\beta\rho}] \quad . \quad (5.14)$$

The result (5.10) was established in FTSPD (Ref 2.) using functional methods and ND1 (Ref. 1) using algebraic methods.

We conveniently introduce the set of cumulants:

$$G_{\alpha\beta\dots\delta} = \frac{\delta}{\delta H_\alpha} \frac{\delta}{\delta H_\beta} \dots \frac{\delta}{\delta H_\delta} W[H] \quad (5.15)$$

to obtain the functional Taylor series expansion (5.13) as,

$$\Delta W[H] = \sum_\alpha F_\alpha G_\alpha + \sum_{\alpha\beta} \frac{1}{2} F_\alpha F_\beta G_{\alpha\beta} + \sum_{\alpha\beta\gamma} \frac{1}{3!} F_\alpha F_\beta F_\gamma G_{\alpha\beta\gamma} + \dots \quad (5.16)$$

Clearly, in this form ΔW is treated as a functional correlation functions G_α , $G_{\alpha\beta}$, $G_{\alpha\beta\gamma}$, ... and irreducible vertex functions.

We established in FTSPD a dynamic generalization of the static Ornstein-Zernike relation[19] given by eqn.(59) there. Inserting Eq.(5.16) in eqn. (5.15) we obtain

$$G_{\alpha\beta} = \frac{\delta}{\delta H_\beta} G_\alpha = \mathcal{G}_{\alpha\beta} + \sum_{\gamma} c_{\alpha\gamma} G_{\gamma\beta} \quad (5.17)$$

where $\mathcal{G}_{\alpha\beta}$ is given by Eq.(2.23) and is related to the selfs- correlation function:

$$\mathcal{G}_{\alpha\beta} = \text{Tr} \phi_\alpha \phi_\beta e^{H \cdot \phi + \Delta W} \quad (5.18)$$

The memory function[20], or dynamic direct correlation function in eqn. (5.17) is obtained as

$$c_{\alpha\beta} = \text{Tr} \phi_\alpha e^{H \cdot \phi + \Delta W} \frac{\delta}{\delta G_\beta} \Delta W. \quad (5.19)$$

Since ΔW can be treated as a functional of G_i we see at this stage that we have available a self-consistent theory. If we define the matrix-inverses

$$\sum_{\delta} \Gamma_{\alpha\delta} G_{\delta\beta} = \delta_{\alpha\beta} \quad (5.20)$$

$$\sum_{\delta} \gamma_{\alpha\delta} \mathcal{G}_{\delta\beta} = \delta_{\alpha\beta} \quad (5.21)$$

then the two-point vertex is given without approximation by

$$\Gamma_{\alpha\beta} = \gamma_{\alpha\beta} + K_{\alpha\beta} \quad (5.22)$$

where

$$K_{\alpha\beta} = - \sum_{\delta} \gamma_{\alpha\delta} \mathcal{C}_{\delta\beta} \quad (5.23)$$

is the collective contribution to the vertex function $\Gamma_{\alpha\beta}$. From eqn. (5.19) it follows that we can obtain the $c_{\alpha\beta}$ and hence $K_{\alpha\beta}$ as an expansion in the interaction potential by expressing ΔW in a similar series

$$\Delta W = \Delta W^{(1)} + \Delta W^{(2)} + \Delta W^{(3)} + \dots \quad (5.24)$$

B. Collective part of vertex function at First-order

Going to first order in perturbation theory we have

$$\Delta W^{(1)} = \sum_{\alpha} F_{\alpha} G_{\alpha} \quad . \quad (5.25)$$

Then we need to compute

$$f_{\alpha}^{(1)} = \frac{\delta}{\delta G_{\alpha}} \Delta W^{(1)} = F_{\alpha} \quad (5.26)$$

which goes back into Eq.(5.19) giving the result

$$c_{\alpha\beta} = \text{Tr} \phi_{\alpha} F_{\beta} e^{\Delta W} = \sum_{\delta} \mathcal{G}_{\alpha\delta} \sigma_{\delta\beta} \quad . \quad (5.27)$$

Putting Eq.(5.27) into Eq.(5.23) and using Eq.(5.21) gives the very simple result

$$K_{\alpha\beta}^{(1)} = -\sigma_{\alpha\beta} \quad . \quad (5.28)$$

This result satisfies the FDR discussed below in a trivial way:

$$K_{\rho B}^{(1)} = K_{B\rho}^{(1)} = -\beta V(q) \quad (5.29)$$

while

$$K_{BB}^{(1)} = -\frac{2}{\beta\omega} \text{Im} K_{B\rho}^{(1)} = 0 \quad . \quad (5.30)$$

C. Collective part of the vertex function at Second Order

The collective part K of the two-point vertex function Γ , at second order is denoted as $K^{(2)}$ and is obtained by determining $f_{\alpha}^{(2)}$, defined as

$$f_{\alpha}^{(2)} = \frac{\delta}{\delta G_{\alpha}} \Delta W^{(2)} \quad (5.31)$$

at second order. We have

$$\Delta W^{(2)} = \frac{1}{2} \sum_{\alpha\beta} F_{\alpha} F_{\beta} G_{\alpha\beta} \quad (5.32)$$

and

$$f_{\alpha}^{(2)} = \frac{\delta \Delta W^{(2)}}{\delta G_{\alpha}} = \frac{1}{2} \sum_{\beta\delta} F_{\beta} F_{\delta} \frac{\delta G_{\beta\delta}}{\delta G_{\alpha}} \quad . \quad (5.33)$$

We then have the standard functional manipulations,

$$\frac{\delta}{\delta G_\delta} G_{\alpha\beta} = - \sum_{\nu\eta} G_{\alpha\nu} \frac{\delta G_{\nu\eta}^{-1}}{\delta G_\delta} G_{\eta\beta} = - \sum_{\nu\eta} G_{\alpha\nu} G_{\beta\eta} \Gamma_{\nu\eta\gamma} \quad (5.34)$$

where in writing the last equation we introduce the three-point vertex

$$\Gamma_{\alpha\beta\gamma} = \frac{\delta}{\delta G_\gamma} \Gamma_{\alpha\beta} \quad . \quad (5.35)$$

Putting this back into Eq.(5.33) gives

$$f_\alpha^{(2)} = \frac{\delta \Delta W^{(2)}}{\delta G_\alpha} = -\frac{1}{2} \sum_{\nu\mu\kappa\delta} F_\nu F_\mu G_{\nu\kappa} G_{\mu\delta} \Gamma_{\kappa\delta\alpha} \quad . \quad (5.36)$$

This in turn goes into Eq.(5.19) and

$$\begin{aligned} c_{\alpha\beta}^{(2)} &= -\frac{1}{2} \sum_{\kappa\gamma\delta\eta} \text{Tr} \phi_\alpha e^{\Delta W} F_\kappa F_\gamma G_{\kappa\delta} G_{\gamma\eta} \Gamma_{\delta\eta\beta} \\ &= -\frac{1}{2} \sum_{\delta\nu\kappa\eta\pi\varrho\theta} \mathcal{G}_{\alpha\delta\nu} \sigma_{\delta\eta} \sigma_{\nu\pi} G_{\eta\varrho} G_{\pi\theta} \Gamma_{\varrho\theta\beta} \end{aligned}$$

where

$$\mathcal{G}_{\alpha\beta\delta} = \text{Tr} \phi_\alpha \phi_\beta \phi_\delta e^{\Delta W} \quad (5.37)$$

is a three-point self-correlation involving the products of three single particle quantities $\phi_\alpha^{(i)}$, $\phi_\beta^{(i)}$ and $\phi_\delta^{(i)}$. The second order contributions to the two-point vertex function $\Gamma_{\alpha\beta}$ can be written more symmetrically in terms of the three-point self-vertex $\gamma_{\alpha\beta\delta}$. The latter is defined as

$$\mathcal{G}_{\alpha\beta\gamma} = - \sum_{\alpha'\beta'\delta'} \mathcal{G}_{\alpha\alpha'} \mathcal{G}_{\beta\beta'} \mathcal{G}_{\delta\delta'} \gamma_{\alpha'\beta'\delta'} \quad . \quad (5.38)$$

Then the collective part of the two-point-vertex is given at second-order by

$$\begin{aligned} K_{\alpha\beta}^{(2)} &= -\frac{1}{2} \sum_{\nu\nu'\mu\mu'\delta\delta'\eta\eta'\pi\pi'} \gamma_{\alpha\nu} \mathcal{G}_{\nu\nu'} \mathcal{G}_{\mu\mu'} \mathcal{G}_{\delta\delta'} \gamma_{\nu'\mu'\delta'} \sigma_{\mu\eta'} \sigma_{\delta\pi'} G_{\eta'\eta} G_{\pi'\pi} \Gamma_{\beta\eta\pi} \\ &= -\frac{1}{2} \sum_{\nu\nu'\mu\mu'\delta\delta'\eta\eta'\pi\pi'} \gamma_{\alpha\nu'\delta'} \mathcal{G}_{\nu\nu'} \mathcal{G}_{\delta\delta'} \sigma_{\nu\eta} \sigma_{\delta\pi} G_{\eta\eta'} G_{\pi\pi'} \Gamma_{\eta'\pi'\beta} \\ &= -\frac{1}{2} \sum_{\nu\mu\delta\eta\pi} \gamma_{\alpha\nu\delta} \bar{G}_{\nu\mu} \bar{G}_{\delta\eta} \Gamma_{\mu\eta\beta} \end{aligned} \quad (5.39)$$

where in writing the last equality we have introduced the self-dressed propagator

$$\bar{G}_{\alpha\beta} = \sum_{\nu\delta} \mathcal{G}_{\alpha\nu} \sigma_{\nu\delta} G_{\delta\beta} \quad . \quad (5.40)$$

In Appendix C we demonstrate that $\bar{G}_{\alpha\beta}$ is a FDRMP with the same generic properties as $G_{\alpha\beta}$.

From its definition the collective part of the two-point vertex can be constructed to be symmetric and the second-order contribution can be written in the symmetric form:

$$\begin{aligned} K_{\alpha\beta}^{(2)} &= -\frac{1}{2} \sum_{\nu\delta\eta\theta} [\gamma_{\alpha\nu\delta} \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} \Gamma_{\eta\theta\beta} + \gamma_{\beta\nu\delta} \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} \Gamma_{\eta\theta\alpha}] \\ &= -\frac{1}{2} \sum_{\nu\delta\eta\theta} [\gamma_{\alpha\nu\delta} \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} \Gamma_{\eta\theta\beta} + \Gamma_{\alpha\nu\delta} \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} \gamma_{\eta\theta\beta}] \\ &= -\frac{1}{2} \sum_{\nu\delta\eta\theta} \left[(\Gamma_{\alpha\nu\delta} + \gamma_{\alpha\nu\delta}) \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} (\Gamma_{\eta\theta\beta} + \gamma_{\eta\theta\beta}) - \Gamma_{\alpha\nu\delta} \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} \Gamma_{\eta\theta\beta} \right. \\ &\quad \left. - \gamma_{\alpha\nu\delta} \bar{G}_{\nu\eta} \bar{G}_{\delta\theta} \gamma_{\eta\theta\beta} \right], \end{aligned} \quad (5.41)$$

We look at the properties of $K_{\alpha\beta}^{(2)}$ in detail in section VI.

D. Single-particle contribution to vertex function

Besides the collective contribution to the vertex function $\Gamma_{\alpha\beta}$, we have the single-particle contribution. Following the discussion in MMS[7] we find that the single-particle contribution $\gamma_{\alpha\beta}$ to the memory function $\Gamma_{\alpha\beta}$ at the second order is given by

$$\gamma_{\alpha\beta}^{(2)} = - \sum_{\nu\delta\eta\theta} \gamma_{\alpha\nu\delta} \tilde{G}_{\nu\eta} \mathcal{G}_{\delta\theta} \gamma_{\eta\theta\beta} \quad . \quad (5.42)$$

To avoid ambiguity the collective contribution obtained in eqn. (5.41) is to be denoted as $K_{\alpha\beta}^{(2)}$. The vertex function $\gamma_{\alpha\beta\delta}$ is defined by Eq.(5.38) and two-point single-particle propagator \mathcal{G} is defined in (5.18). The quantity $\tilde{G}_{\alpha\beta}$ is another dressed propagator similar to $\bar{G}_{\alpha\beta}$ defined in eqn. (5.40) corresponding to the collective contribution $K_{\alpha\beta}$. $\tilde{G}_{\alpha\beta}$ is defined as

$$\tilde{G}_{\alpha\beta} = \sum_{\nu\delta\eta\pi} \mathcal{G}_{\alpha\nu} \sigma_{\nu\delta} G_{\delta\eta} \sigma_{\eta\pi} \mathcal{G}_{\pi\beta} \quad (5.43)$$

In Appendix C we show that like the set $\{G^{(0)}, G, \mathcal{G}, \bar{G}\}$, the matrix \tilde{G} also satisfies the property of being a FDRMP.

VI. ONE-LOOP SELF-ENERGY

A. General Structure of One-loop Self-energies

Generally the self energy matrix Σ is defined in terms of the Dyson equation involving inverse of the matrix of the two-point function as follows

$$\Gamma \equiv G^{-1} = G_0^{-1} - \Sigma \quad . \quad (6.1)$$

In the above equation G_0^{-1} refers to the zeroth order vertex functions. In field theoretic models this zeroth order theory stands for the gaussian level theories which usually refers to the trivial case *e.g.*, linear dynamics of fluctuations. In the present context the zeroth order (in interaction potentials) theory refers to the non interacting system. The self energy Σ will be therefore defined here by subtracting the zeroth order and the first order contributions from the full vertex function

$$-\Sigma = \Gamma - \Gamma^{(0)} - \Gamma^{(1)} \quad . \quad (6.2)$$

At the one-loop level the self energy is a sum of collective and single particle contributions respectively denoted by K and γ ,

$$-\Sigma = \gamma^{(2)} + K^{(2)} + \mathcal{O}(3) \quad , \quad (6.3)$$

where the superscript (2) indicates the second order in in the interaction potential and $\mathcal{O}(3)$ stands for higher order contributions. The single particle and the collective contributions are respectively expressed as,

$$\gamma^{(2)} = -\frac{\gamma}{2}\tilde{G}\mathcal{G}\gamma = -\frac{1}{4}\gamma \left[(\tilde{G} + \mathcal{G})(\mathcal{G} + \tilde{G}) - \tilde{G}\tilde{G} - \mathcal{G}\mathcal{G} \right] \gamma \quad (6.4)$$

$$\begin{aligned} K^{(2)} &= -\frac{1}{4}(\gamma\bar{G}\bar{G}\Gamma + \Gamma\bar{G}\bar{G}\gamma) \\ &= -\frac{1}{4}(\gamma + \Gamma)\bar{G}\bar{G}(\gamma + \Gamma) + \frac{1}{4}\gamma\bar{G}\bar{G}\gamma + \frac{1}{4}\Gamma\bar{G}\bar{G}\Gamma \quad . \end{aligned} \quad (6.5)$$

In order to further analyze the one-loop results for γ and K obtained above we introduce the generalized functional $\Pi[E, A]$ of a two-point propagator E and three-point vertex A as

$$\Pi[A, E] = -\frac{1}{2}EAAE \quad . \quad (6.6)$$

The one-loop contributions to the different elements of the self energy Σ matrix defined in eqn. (6.3) are now expressed in terms of those of matrix Π . We focus on the $\alpha\mu$ -th element ($\alpha\mu \in \{\rho, B\}$) of the matrix Π :

$$\Pi_{\alpha\mu}[A, E](-1) = - \sum_{\kappa, \nu, \sigma, \delta} \frac{1}{2} \int d2d3 E_{\alpha\kappa\nu}^*(123) \delta(1+2+3) A_{\kappa\sigma}(2) A_{\nu\delta}(3) E_{\mu\sigma\delta}(123) \quad (6.7)$$

where A is a FDRMP and E is a three-point vertex that satisfies the FDR of section III. In the above definition for Π , the propagator A is from the set $\{G^{(0)}, \mathcal{G}, G, \bar{G}, \tilde{G}\}$ and the three-point vertices E include $\{\gamma^{(0)}, \gamma, \Gamma\}$. In terms of the functional $\Pi[A, E]$, the single-particle and collective contributions respectively given by eqns. (6.4) and (6.5) are obtained in the form:

$$\gamma^{(2)} = \frac{1}{2}\Pi[(\mathcal{G} + \tilde{G}), \gamma] - \frac{1}{2}\Pi[\mathcal{G}, \gamma] - \frac{1}{2}\Pi[\tilde{G}, \gamma] \quad , \quad (6.8)$$

$$K^{(2)} = \frac{1}{2}\Pi[\bar{G}; \gamma + \Gamma] - \frac{1}{2}\Pi[\bar{G}; \gamma] - \frac{1}{2}\Pi[\bar{G}; \Gamma] \quad . \quad (6.9)$$

Next we consider the fluctuation-dissipation symmetry of the single particle and collective contributions to the self energy Σ at one loop level.

B. Partial Self-energies and the FDT

We investigate the existence of a fluctuation-dissipation theorem (FDT) satisfied by the partial self-energy in Fourier space. We first separate $\Pi[A, E]$ into its components, the nonzero choices for the indices κ, ν, σ , and δ . There are nine contributions to $\Pi_{BB}[A, E]$, five contributions to $\Pi_{\rho B}[A, E]$ and five contributions to $\Pi_{\rho\rho}[A, E]$. In order to better organize the algebra we replace all $A_{\rho\rho}$ internal lines using the FDR:

$$A_{\rho\rho}(q, \omega) = \frac{A_{B\rho}(q, \omega) - A_{\rho B}(q, \omega)}{i\beta\omega} . \quad (6.10)$$

For treating the two-point vertices we introduce a set of simplifying relations which are generalizations of eqns. (3.70)-(3.72) in terms of the generalized vertex function E :

$$\phi_i = i\beta\omega_i E_{B_i\rho_j\rho_k}, \quad (6.11)$$

$$\psi_i = \beta^2\omega_j\omega_k E_{\rho_i B_j B_k}, \quad (6.12)$$

$$P = -i\beta^3\omega_i\omega_j\omega_k E_{B_i B_j B_k} . \quad (6.13)$$

where we have explicitly indicated in the first term on LHS the factor of β , which was earlier absorbed in the definition of the frequency ω . The quantities ϕ_i , ψ_i and P defined above in terms of the generalized vertex function $E \in \{\Gamma, \gamma, \gamma^{(0)}\}$ satisfies the same set of identities given by eqns. (3.73) and (3.74). After considerable algebra we find

$$\begin{aligned} & i\omega_1 \Pi_{BB}[A, E] + \Pi_{\rho B}[A, E] - \Pi_{B\rho}[A, E] \\ &= -\frac{i}{D} \int d^2d^3 \left[\left\{ A_{B\rho}(2) A_{B\rho}(3) C_1^* + A_{\rho B}(2) A_{\rho B}(3) C_1 \right\} \right. \\ & \quad \left. - \left\{ A_{B\rho}(2) A_{\rho B}(3) C_2^* + A_{\rho B}(2) A_{B\rho}(3) C_2 \right\} \right] \delta(1+2+3) . \end{aligned} \quad (6.14)$$

We have defined the quantities C_1 , C_2 , and D respectively as,

$$C_1 = \phi_1^*(\phi_1 + \phi_2 + \phi_3 + \psi_1 + \psi_2 + \psi_3 + P) \quad (6.15)$$

$$C_2 = (\phi_1 + \phi_2 + \psi_3)(\phi_1^* + \psi_2^* + \phi_3^*) - \phi_2\phi_3^* \quad (6.16)$$

$$D = \beta^3\omega_1\omega_2\omega_3 . \quad (6.17)$$

Using the identities (3.73) and (3.74) it is straightforward to show that C_1 and C_2 both vanishes, obtaining a relation similar to (3.34) to hold with the elements of the generalized matrix Π :

$$\tilde{\mathcal{F}}[\Pi] \equiv -i\beta\omega_1 \Pi_{BB}(1) + \Pi_{B\rho}(1) - \Pi_{\rho B}(1) = 0 . \quad (6.18)$$

In the above equation and in what follows the functional dependence of $\Pi[E, A]$ on the vertex E and correlation A are not explicitly shown in the RHS to avoid cluttering. In Section III the full two-point vertex function matrix Γ_{ij} was shown to be a FDRMP with the result (3.48). We have demonstrated here that the self energy matrix $\Pi_{\alpha\beta}[E, A]$ expressed in terms of the generalized vertex $E_{\alpha\beta\delta}$ and correlation functions $A_{\beta\delta}$, are also FDRMP. Since $\tilde{\mathcal{F}}$ defined in eqn. (3.35) is linear, using eqn. (6.18) in the definitions (6.5) and (6.4) respectively, it follows that

$$\tilde{\mathcal{F}} [K^{(2)}] = 0 \quad , \quad (6.19)$$

$$\tilde{\mathcal{F}} [\gamma^{(2)}] = 0 \quad . \quad (6.20)$$

Hence both the collective and single-particle one loop level contributions to the self energy respectively denoted by K_{ij} and γ_{ij} are FDRMP.

After making full use of the vertex identities we find that the self energy matrix element $\Pi_{B\rho}(-1)$ is the sum of two pieces

$$-2\Pi_{B\rho}(-1) = i \int \frac{d^2d^3}{D} \delta(1+2+3) \left[(\phi_1^*)^2 A_{\rho B}(2) A_{\rho B}(3) + (\phi_2)^2 A_{\rho B}(2) A_{B\rho}(3) \right] \quad . \quad (6.21)$$

Notice that this self-energy depends only on the three-point vertex functions with one B label; $E_{B\rho\rho}$ and $E_{\rho B\rho}$ respectively as follows from eqn. (6.11).

1. Separation into High and Low Frequency Components

We will be interested in low-frequency-long-time phenomena. We identify the low-frequency contribution to the vertex using the Vertex Theorem (see appendix B) as :

$$E_{\rho B\rho}(0, 0, 0; q_1, k_2, -q_1 - k_2) = \gamma_{\rho\rho\rho}(q_1, k_2, -q_1 - k_2) \quad (6.22)$$

where $\gamma_{\rho\rho\rho}$ is the static 3-point vertex and write the frequency dependent vertex functions as,

$$\begin{aligned} E_{\rho B\rho}(\omega_1, \omega_2, \omega_3; q_1, k_2, -q_1 - k_2) &= \gamma_{\rho\rho\rho}(q_1, k_2, -q_1 - k_2) \\ &+ \Delta_{\rho B\rho}(\omega_1, \omega_2, \omega_3; q_1, k_2, -q_1 - k_2) \quad . \end{aligned} \quad (6.23)$$

The one loop contribution denoted by $\Pi[E, A]$ is split in to a low frequency part $\hat{\Pi}$ (for $\omega \rightarrow 0$) and a high frequency part $\tilde{\Pi}$

$$\Pi_{B\rho}(1) = \hat{\Pi}_{B\rho}(1) + \tilde{\Pi}_{B\rho}(1) \quad (6.24)$$

where $\hat{\Pi}_{B\rho}(1)$ is the contribution with $E_{\rho B\rho}$ replaced by its static limit value. After a large amount of algebra we find the low frequency contribution $\hat{\Pi}(1)$ is reduced to the form,

$$\begin{aligned} \hat{\Pi}_{B\rho}(1) &= \frac{1}{2\beta} \hat{\mathcal{O}} \bar{A}_{\rho\rho}(k_2) \bar{A}_{\rho\rho}(k_3) [\gamma_{\rho\rho\rho}(q_1, k_2, k_3)]^2 \\ &+ \frac{\omega_1}{2\beta} \hat{\mathcal{O}} \int \frac{dx}{2\pi} \int \frac{dy}{2\pi} [\gamma_{\rho\rho\rho}(q_1, k_2, k_3)]^2 \frac{A_{\rho\rho}(x, k_2) A_{\rho\rho}(y, k_3)}{-\omega_1 + x + y - i\eta} , \end{aligned} \quad (6.25)$$

where we have defined the operator $\hat{\mathcal{O}}$ as

$$\hat{\mathcal{O}} = \int \frac{d^d k_2}{(2\pi)^d} \int \frac{d^d k_3}{(2\pi)^d} \delta(q_1 - k_2 - k_3) \quad . \quad (6.26)$$

The static ($\omega \rightarrow 0$) contribution $\hat{\Pi}(1)$ is given by

$$\hat{\Pi}_{B\rho}(q_1, 0) = \frac{1}{2\beta} \hat{\mathcal{O}} \bar{A}_{\rho\rho}(k_2) \bar{A}_{\rho\rho}(k_3) [\gamma_{\rho\rho\rho}(q_1, k_2, k_3)]^2 \quad . \quad (6.27)$$

Since the low-frequency part $\hat{\Pi}$ is itself a FDRMP, the BB element of the $\hat{\Pi}$ matrix is given by

$$\begin{aligned} \hat{\Pi}_{BB}(1) &= -\frac{2\beta^{-1}}{\omega_1} \text{Im} \left[\hat{\Pi}_{B\rho}(1) \right] \\ &= -\frac{1}{2\beta^2} \hat{\mathcal{O}} \int \frac{dx}{2\pi} [\gamma_{\rho\rho\rho}(q_1, k_2, k_3)]^2 A_{\rho\rho}(x, k_2) A_{\rho\rho}(\omega_1 - x, k_3) \quad . \end{aligned} \quad (6.28)$$

Notice that it follows directly from eqns. (6.25) and (6.28) that the sum rule

$$\hat{\Pi}_{B\rho}(q, 0) = - \int \frac{d\omega}{2\pi} \beta \hat{\Pi}_{BB}(q, \omega) \quad (6.29)$$

is satisfied. The high-frequency contribution $\tilde{\Pi}_{B\rho}(\omega)$ to the self energy satisfy

$$\frac{\tilde{\Pi}_{B\rho}(1)}{\hat{\Pi}_{B\rho}(1)} \approx \omega_1 \quad (6.30)$$

as $\omega_1 \rightarrow 0$, and hence does not contribute to the slow dynamics of the system. A key result in this work is the theorem that $\Gamma_{B\rho\rho}$ reduces to the static three-point vertex in the low-frequency limit.

C. Original One loop Problem

We now focus on the one loop contributions for collective and single-particle self-energies respectively denoted by $K_{\alpha\mu}$ and $\gamma_{\alpha\mu}$ which were introduced earlier in section VI. For the collective contribution K the appropriate three-point vertex functions are $E \equiv \{(\gamma + \Gamma), \gamma, \Gamma\}$, while the correlation function $A \equiv \bar{G}$. The collective contribution is divided in to two parts as

$$K_{\alpha\mu}(1) = \hat{K}_{\alpha\mu}(1) + \tilde{K}_{\alpha\mu}(1) \quad (6.31)$$

where we have followed the same notations in terms of bar and tilde on the respective terms as given in eqn. (6.24) in indicating the low and high frequency components. Since $K_{\alpha\mu}$ is a FDRMP, we obtain from eqn. (6.5) and (6.28), the low frequency part of the collective contribution $K_{BB}^{(2)}$ as

$$\begin{aligned} \hat{K}_{BB}^{(2)}(1) &= -\frac{2\beta^{-1}}{\omega_1} \text{Im} \hat{K}_{B\rho}^{(2)}(1) \\ &= -\frac{1}{\beta^2} \hat{\mathcal{O}} \int \frac{dx}{2\pi} \bar{G}_{\rho\rho}(x, k_2) \int \frac{dy}{2\pi} \bar{G}_{\rho\rho}(y, k_3) \pi \delta(\omega_1 - x - y) \\ &\quad \times \left[\frac{1}{2} \{(\gamma_{\rho\rho\rho} + \Gamma_{\rho\rho\rho})\}^2 - \frac{1}{2} (\gamma_{\rho\rho\rho})^2 - \frac{1}{2} (\Gamma_{\rho\rho\rho})^2 \right] \\ &= -\frac{1}{2\beta^2} \hat{\mathcal{O}} \int \frac{dx}{2\pi} \bar{G}_{\rho\rho}(x, k_2) \bar{G}_{\rho\rho}(\omega_1 - x, k_3) \gamma_{\rho\rho\rho} \Gamma_{\rho\rho\rho} \quad . \end{aligned} \quad (6.32)$$

For the single particle contribution $\gamma_{\alpha\mu}$ the appropriate three-point vertex functions are $E \equiv \gamma$, while the correlation function $A \equiv \{(\mathcal{G} + \tilde{G}), \mathcal{G}, \tilde{G}\}$. We obtain from eqn. (6.4) the single particle contribution as,

$$\hat{\gamma}_{BB}^{(2)}(1) = -\frac{1}{2\beta^2} \hat{\mathcal{O}} \int \frac{dx}{2\pi} \tilde{G}_{\rho\rho}(x, k_2) \mathcal{G}_{\rho\rho}(\omega_1 - x, k_3) \gamma_{\rho\rho\rho}^2 \quad . \quad (6.33)$$

Eqns. (6.32) and (6.33) are approximations for the second-order collective and single particle contributions to the frequency dependent self energy Σ **including** vertex corrections. We write for the respective collective and single particle contributions in the time regime

$$\hat{K}_{BB}^{(2)}(q_1, t_1) = -\frac{1}{2\beta^2} \hat{\mathcal{O}} \bar{G}_{\rho\rho}(t_1, k_2) \bar{G}_{\rho\rho}(t_1, k_3) \gamma_{\rho\rho\rho} \Gamma_{\rho\rho\rho} \quad . \quad (6.34)$$

and

$$\hat{\gamma}_{BB}^{(2)}(q_1, t_1) = -\frac{1}{2\beta^2} \hat{\mathcal{O}} \tilde{G}_{\rho\rho}(t_1, k_2) \mathcal{G}_{\rho\rho}(t_1, k_3) \gamma_{\rho\rho\rho}^2 \quad . \quad (6.35)$$

In the low-frequency limit we assume, as we show self-consistently, $\hat{K}_{BB}^{(2)}(q_1, z_1)$ grows arbitrarily large as the frequency z_1 goes to zero. Since $\hat{\gamma}_{BB}^{(2)}(q_1, 0)$ is regular we can drop the single-particle contribution to the memory function in the kinetic equation.

In the expression given in Eq. (6.34) for the memory function, the implication of having the full vertex function $\Gamma_{\rho\rho\rho}$ on the dynamic behavior of the fluid has been ignored. To lowest order in the interaction potential, the vertex $\Gamma_{\rho\rho\rho}$ is approximated by the corresponding quantity $\gamma_{\rho\rho\rho}$ of the non-interacting theory which is the case for the ideal gas. Since the vertex functions are only involved here (see Appendix B) in the $\omega \rightarrow 0$ limit and hence they appears like a static quantity. So far these static approximations are not directly connected with HNC or PY approximations. We discuss the role of vertex corrections elsewhere. We have then

$$\Gamma_{\rho\rho\rho} = -\gamma_{\rho\rho\rho} = -\frac{1}{\bar{\rho}^2} \quad . \quad (6.36)$$

We also assume consistent with the long-time approximation that for low-frequencies

$$G_{\rho\rho}(q, \omega) \gg G_{\rho B}(q, \omega) \quad (6.37)$$

and in $\bar{G}_{\rho\rho}(q, \omega)$ the $G_{\rho\rho}(q, \omega)$ term dominates and its coefficient can be replaced by its $\omega = 0$ value. Using the definitions $G_{\rho\rho}(q, \omega) = \bar{\rho} S(q) F(q, \omega)$ we find

$$\begin{aligned} \bar{G}_{\rho\rho}(q, \omega) &= \beta V(q) \bar{\rho} G_{\rho\rho}(q, \omega) \\ &= \tilde{V}(q) \bar{\rho} S(q) F(q, \omega) \quad , \end{aligned} \quad (6.38)$$

where we have denoted $\tilde{V} = \bar{\rho} \beta V(q)$ as the scaled potential. From the above definitions we obtain the relation $\tilde{F}(q, t) = \tilde{V}(q) S(q) F(q, t)$.

We can then write in the long time regime the simple result

$$K_{BB}^{(2)}(q, t) = -\frac{1}{2\bar{\rho}^2\beta^2} \int \frac{d^d k}{(2\pi)^d} \tilde{F}(q-k, t) \tilde{F}(k, t) \quad (6.39)$$

We need the Fourier-Laplace transform of Eq.(4.33) to go back in the low-frequency form of the kinetic equation. After some simple rearrangements we obtain our main result from Eq.(4.35)

$$\frac{F(q, z)}{1 - zF(q, z)} = -i\frac{S(q)}{2\bar{\rho}} \int_0^\infty dt e^{izt} \int \frac{d^d k}{(2\pi)^d} \tilde{F}(q-k, t) \tilde{F}(k, t) \quad (6.40)$$

This is a highly nonlinear equation for $F(q, t)$. This result is identical, after reconciling notation, to the result in Ref. [6] for Smoluchowsky dynamics. As in SM we can carry out an analytic treatment of Eq.(4.33) by expanding

$$F(q, z) = \frac{f(q)}{z} + \psi(q, z) \quad (6.41)$$

for $z \rightarrow 0$ and $\lim_{z \rightarrow 0} z\psi(q, z) \rightarrow 0$ while $\lim_{z \rightarrow 0} \psi(q, z) \rightarrow \infty$. $f(q)$ is the nonergodicity parameter with the interpretation

$$\lim_{t \rightarrow \infty} G_{\rho\rho}(q, t) = f(q)\bar{\rho}S(q) \quad . \quad (6.42)$$

In the small z or long time limit the eqn. (6.40) reduces to an integral equation in terms of the nonergodicity parameters $f(q)$ of the form

$$\frac{f(q)}{1 - f(q)} = \frac{S(q)}{2\bar{\rho}} \int \frac{d^d k}{(2\pi)^d} S(q-k) \tilde{V}(q-k) f(q-k) S(k) \tilde{V}(k) f(k) \quad . \quad (6.43)$$

This equation must be supplemented with an equation connecting the potential and the static structure factor. At second order in the potential we have

$$S^{-1}(q) - 1 = \tilde{V}(q) - \frac{1}{2\bar{\rho}} \int \frac{d^d k}{(2\pi)^d} \tilde{V}(q-k) S(q-k) \tilde{V}(k) S(k) \quad (6.44)$$

We choose to fix $S(q)$ and solve for the pseudo-potential $\tilde{V}(q)$. The question of a pseudo-potential is discussed in detail in SDENE. We can determine $\tilde{V}(q)$ once we have a form for the

static structure factor. Typically we have used the exact solution of the approximate Percus-Yevick[21] equation for hard spheres. One can then carry out the explicit determination of the quantities characteristic of the ENE transition.

The above integral equations for $f(q)$ is solved over a grid to obtain the nonzero solutions using iterative methods. The density at which the trivial solution $f(q) = 0$ changes to a nonzero set of nonergodicity parameter values marks the location of the ideal ENE transition in the dense liquid approaching from the liquid side.

In Fig. C we display the static structure factor for the hard sphere liquid as obtained from Percus-Yevick[21] equations with Verlet-Weiss corrections [22] at packing fraction $\eta = 0.62$. Using this $S(k)$ as an input we solve iteratively eqns. (6.44) to obtain the renormalized potential $\tilde{V}(q)$. The wave vector q is chosen over a grid of upper cutoff value $q\sigma = 80$ and having 500 points. Fig. C displays the $\tilde{V}(q)$ obtained with input structure factor of Fig. C.

Next we use this renormalized potential to evaluate the vertex functions in eqn. (6.43) for the nonergodicity parameters $f(q)$. Solving these integral equations iteratively we obtain that the $f(q)$ are vanishing till the critical packing fraction of $\eta = .62$. The corresponding nonergodicity parameters $f(q)$ over the whole wave vector grid is shown in Fig. C. The ENE transition point of the core problem of self-consistent feed back mechanism using Percus-Yevick structure factors with Verlet-Weiss corrections therefore at packing fraction of .62 which is close to the close-packing density.

VII. CONCLUSIONS

We have shown within the pseudo-potential expansion the ENE problem for ND reduces to precisely the same problem for SD. For hard-spheres and using the PY approximation for the structure factor we can solve the approximate ENE problem to find $\eta^* = 0.62$, and the two-step exponents a and b consistent with the results found in SM. This is a nontrivial result and is closely associated with the vertex theorem which relates the low-frequency limit of $\Gamma_{B\rho\rho}$ to the static vertex $\gamma_{\rho\rho\rho}$. While we have proved this result only to second order in perturbation theory, we expect it is more general[13].

We have found simplification of the ND case only in the low-frequency regime. More generally the SD case is considerably simpler as demonstrated in MMS where the zeroth order vertices are very simple.

This result for the dynamic structure factor raises questions about higher-order correlation functions such as $G_{\rho\rho\rho}(123)$. There is good reason to believe that we can make progress in determining this quantity in the low-frequency regime.

Our results here depend on ignoring static-vertex corrections and higher-order loop contributions. We need to investigate these correction to see if the results for the ENE are stable. We see from Eq.(6.34) that the low-frequency dynamics depends not just on the static structure factor, but also on the static three-point vertex.

The theory presented in this work has been set up to deal with what we call the *core* problem in the ND case. The primary motivation here is the determination of the observables involving these core variables $\Phi_0 = (\rho, B)$. The terms core refers to the fact that the Hamiltonian and hence the corresponding MSR action is expressed in terms of only these variables. As already pointed out (see sec. 9 in ND1), by going beyond the core variables, considerations of additional degrees of freedom enter the theory. For example in the ND case, we have a larger phase-space due to the momentum degrees of freedom and there are additional conservation laws for the system. If one extends the set of core variables to include the momentum density, we have all the correlation functions among ρ , B and g in the description. The resulting hierarchical structure in the formal expressions for the correlation functions may change the viability of the ENE transition being discussed here. In this regard it is useful to note the known results of including the momentum density fluctuations in the fluctuating hydrodynamic approach to the problem. The momentum density coupling to the density fluctuations in the hydrodynamic equations give rise to the ergodicity restoring mechanisms [23, 24]. In the case of SD on the other hand, the core problem itself covers essentially all of the degrees of freedom of interest. However the microscopic dynamics in this case is dissipative and the equations of motion for the collective collective variables [25, 26] involve multiplicative noise which may have implications in restoring ergodic behavior in a liquid. The present work demonstrates that in the case of the core problem through a reorganization of the perturbation theory in terms of an effective potential, the ideal ENE transition is pushed to the close pack density for a hard sphere system.

Appendix A: Three point vertex functions

We obtain the FDT relations between the different three-point vertex functions here. For this we use the general definition of the three point correlation functions and the corresponding three-point vertex function as,

$$G_{\alpha_i \mu_j \nu_k} = -G_{\alpha \alpha'}(i) G_{\mu \mu'}(j) G_{\nu \nu'}(k) \Gamma_{\alpha'_i \mu'_j \nu'_k} \quad (\text{A1})$$

where α_i stands for the field α at the point i and so on where everything is in Fourier space. The labels α, μ and ν are respectively taken from the set $\{\rho, B\}$. Using the result that $\Gamma_{\rho_i \rho_j \rho_k} = 0$, we obtain from the formula (A1), the various cumulants are :

$$\begin{aligned} -G_{\rho_i \rho_j \rho_k} &= G_{\rho \rho}(i) G_{\rho \rho}(j) G_{\rho B}(k) \Gamma_{\rho_i \rho_j B_k} + G_{\rho \rho}(i) G_{\rho B}(j) G_{\rho \rho}(k) \Gamma_{\rho_i B_j \rho_k} \\ &+ G_{\rho B}(i) G_{\rho \rho}(j) G_{\rho \rho}(k) \Gamma_{B_i \rho_j \rho_k} + G_{\rho \rho}(i) G_{\rho B}(j) G_{\rho B}(k) \Gamma_{\rho_i B_j B_k} \\ &+ G_{\rho B}(i) G_{\rho \rho}(j) G_{\rho B}(k) \Gamma_{B_i \rho_j B_k} + G_{\rho B}(i) G_{\rho B}(j) G_{\rho \rho}(k) \Gamma_{B_i B_j \rho_k} \\ &+ G_{\rho B}(i) G_{\rho B}(j) G_{\rho B}(k) \Gamma_{B_i B_j B_k} \end{aligned} \quad (\text{A2})$$

$$\begin{aligned} -G_{B_i \rho_j \rho_k} &= G_{B \rho}(i) G_{\rho B}(j) G_{\rho \rho}(k) \Gamma_{\rho_i B_j \rho_k} + G_{B \rho}(i) G_{\rho \rho}(j) G_{\rho B}(k) \Gamma_{\rho_i \rho_j B_k} \\ &+ G_{B \rho}(i) G_{\rho B}(j) G_{\rho B}(k) \Gamma_{\rho_i B_j B_k} \end{aligned} \quad (\text{A3})$$

$$-G_{B_i B_j \rho_k} = G_{B \rho}(i) G_{B \rho}(j) G_{\rho B}(k) \Gamma_{\rho_i \rho_j B_k} \quad . \quad (\text{A4})$$

Substituting the results (A2)-(A4) in eqn. (3.54), we obtain an expansion in terms of the various three-point vertex functions. For example the coefficient of the vertex function $\Gamma_{B_i \rho_j \rho_k}$ is obtained after some trivial but tedious algebra as

$$\begin{aligned} \Gamma_{B_i \rho_j \rho_k} &: i\omega_i G_{\rho B}(i) G_{B \rho}(j) G_{B \rho}(k) + \omega_i \omega_j G_{\rho B}(i) G_{\rho \rho}(j) G_{B \rho}(k) \\ &= i\omega_i G_{\rho B}(i) G_{\rho B}(j) \{G_{B \rho}(k) - i\omega_k G_{\rho \rho}(k)\} = i\omega_i G_{\rho B}(i) G_{\rho B}(j) G_{\rho B}(k) \end{aligned} \quad (\text{A5})$$

In reaching the above result we have used the FDT relation (3.48). Similarly for the other two vertices with one B field, *i.e.*, $\Gamma_{\rho_i B_j \rho_k}$ and $\Gamma_{\rho_i \rho_j B_k}$ are obtained. For the vertex with two B fields, *e.g.*, $\Gamma_{B_i B_j \rho_k}$ we obtain the corresponding coefficient as

$$\begin{aligned} \Gamma_{B_i B_j \rho_k} &: \omega_i \omega_j G_{\rho B}(i) G_{\rho B}(j) G_{B \rho}(k) - i\omega_i \omega_j \omega_k G_{\rho B}(i) G_{\rho B}(j) G_{\rho \rho}(k) \\ &= \omega_i \omega_j G_{\rho B}(i) G_{\rho B}(j) G_{\rho B}(k) \end{aligned} \quad (\text{A6})$$

and similarly we obtain the coefficients of the other two vertices each with two B fields. After organizing the coefficients of the different three-point vertex functions the following result is obtained.

$$i\left(\omega_i\Gamma_{B_i\rho_j\rho_k} + \omega_j\Gamma_{\rho_i B_j\rho_k} + \omega_k\Gamma_{\rho_i\rho_j B_k}\right) - i\omega_i\omega_j\omega_k\Gamma_{B_i B_j B_k} + \left\{\omega_i\omega_j\Gamma_{B_i B_j\rho_k} + \omega_j\omega_k\Gamma_{\rho_i B_j B_k} + \omega_k\omega_i\Gamma_{B_i\rho_j B_k}\right\} = 0 \quad (\text{A7})$$

In reaching the above result we have dropped a nonzero common factor of $G_{\rho B}(i)G_{\rho B}(j)G_{\rho B}(k)$ from the LHS.

Next, consider the relations (3.55) involving three-point cumulants having *two* B fields. Once again substituting the results (A3)-(A4) in eqn. (3.55), we obtain after organizing the coefficients of the different three-point vertex functions the following result.

$$G_{B\rho}(k)\left\{i\omega_k\Gamma_{\rho_i\rho_j B_k}^* + i\omega_i\Gamma_{B_i\rho_j\rho_k} + i\omega_j\Gamma_{\rho_i B_j\rho_k} + \omega_i\omega_j\Gamma_{B_i B_j\rho_k}\right\} = G_{\rho B}(k)\left\{i\omega_i\Gamma_{B_i\rho_j\rho_k} + i\omega_j\Gamma_{\rho_i B_j\rho_k} + i\omega_k\Gamma_{\rho_i\rho_j B_k} + \omega_i\omega_j\Gamma_{B_i B_j\rho_k} + \omega_i\omega_k\Gamma_{B_i\rho_j B_k} + \omega_j\omega_k\Gamma_{\rho_i B_j B_k} - i\omega_i\omega_j\omega_k\Gamma_{B_i B_j B_k}\right\} = 0 \quad . \quad (\text{A8})$$

In obtaining the above result, we have dropped the common factor of $G_{\rho B}(i)G_{\rho B}(j)$ from both sides and used the basic FD relation (3.48). Using eqn. (3.65) in the result (A8) we find that the coefficient of $G_{\rho B}(k)$ vanishes and one has the result

$$i\omega_k\Gamma_{\rho_i\rho_j B_k}^* + i\omega_i\Gamma_{B_i\rho_j\rho_k} + i\omega_j\Gamma_{\rho_i B_j\rho_k} + \omega_i\omega_j\Gamma_{B_i B_j\rho_k} = 0 \quad . \quad (\text{A9})$$

Appendix B: Higher-Order Thermodynamic Sum Rule

1. Three-point Quantities

Let us consider the low frequency behavior of the full three-point cumulant $G_{\rho BB}(123)$. The quantity $\rho(1)$ in $G_{\rho BB}(123)$ can be replaced by an arbitrary function of density as long as each density corresponds to the same time. The three-point vertex $\Gamma_{B\rho\rho}(123)$ is related to the 3-point correlation $G_{\rho BB}$ by the general relation

$$\Gamma_{B\rho\rho}(123) = -\Gamma_{B\rho}(1)\Gamma_{\rho B}(2)\Gamma_{\rho B}(3)G_{\rho BB}(123) \quad . \quad (\text{B1})$$

Among the FDR identities for the 3-point cumulants, we have for the imaginary part of $G_{\rho BB}$ the relation :

$$G''_{\rho BB}(123) = \frac{\beta\omega_2}{2}G'_{\rho\rho B}(123) + \frac{\beta\omega_3}{2}G'_{\rho B\rho}(123) \quad . \quad (\text{B2})$$

Next, look at the definition of the inverse time fourier transform

$$G_{\rho BB}(q; t_1, t_2, t_3) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} e^{-i\omega_1(t_1-t_3)} e^{-i\omega_2(t_2-t_3)} \tilde{G}_{\rho BB}(\omega_1, \omega_2) \quad (\text{B3})$$

where we have introduced

$$G_{\alpha\beta\gamma}(123) = \tilde{G}_{\alpha\beta\gamma}(\omega_1, \omega_2, q_1, q_2, q_3) \delta(\omega_1 + \omega_2 + \omega_3) \quad . \quad (\text{B4})$$

In this notation we do not always write the third frequency entry since it is implied:

$$\tilde{G}_{\alpha\beta\gamma}(\omega_1, \omega_2) = \tilde{G}_{\alpha\beta\gamma}(\omega_1, \omega_2, -\omega_1 - \omega_2) \quad (\text{B5})$$

and the wave number dependence has been suppressed. Setting $t_2 = t_3$ in eqn. (B3) we obtain,

$$G_{\rho BB}(t_1, t_2, t_2) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} e^{-i\omega_1(t_1-t_2)} \tilde{G}_{\rho BB}(\omega_1, \omega_2) \quad . \quad (\text{B6})$$

This vanishes for $t_2 > t_1$. This is consistent with

$$\int \frac{d\omega_2}{2\pi} \tilde{G}_{\rho BB}(\omega_1, \omega_2) \quad (\text{B7})$$

being analytic in the UHP for ω_1 . Assuming $\tilde{G}_{\rho BB}(\omega_1, \omega_2)$ is analytic in the UHP for ω_1 we can write a dispersion relation

$$\tilde{G}_{\rho BB}(\omega_1, \omega_2) = \int \frac{d\bar{\omega}}{\pi} \frac{\tilde{G}''_{\rho BB}(\bar{\omega}, \omega_2)}{\bar{\omega} - \omega_1 - i\eta} \quad . \quad (\text{B8})$$

Putting Eq.(B4) in Eq.(B2) gives

$$\tilde{G}''_{\rho BB}(\omega_1, \omega_2) = \frac{\beta\omega_2}{2} \tilde{G}'_{\rho\rho B}(\omega_1, \omega_2) - \frac{\beta(\omega_1 + \omega_2)}{2} \tilde{G}'_{\rho B\rho}(\omega_1, \omega_2) \quad . \quad (\text{B9})$$

Putting Eq.(B9) in Eq.(B8) gives

$$\tilde{G}_{\rho BB}(\omega_1, \omega_2) = \int \frac{d\bar{\omega}}{2\pi} \frac{\beta\omega_2 \tilde{G}'_{\rho\rho B}(q; \bar{\omega}, \omega_2) - \beta[\bar{\omega} + \omega_2] \tilde{G}'_{\rho B\rho}(\bar{\omega}, \omega_2)}{\bar{\omega} - \omega_1 - i\eta} . \quad (\text{B10})$$

Letting ω_1 and ω_2 go to zero gives

$$\tilde{G}_{\rho BB}(0, 0) = -\beta \int \frac{d\bar{\omega}}{2\pi} \tilde{G}'_{\rho B\rho}(\bar{\omega}, 0) \quad (\text{B11})$$

We have a FDR identity

$$\tilde{G}''_{\rho B\rho}(\omega_1, \omega_2) = \frac{1}{2}\beta\omega_2 \tilde{G}_{\rho\rho\rho}(\omega_1, \omega_2) \quad (\text{B12})$$

which tells us that

$$\tilde{G}''_{\rho B\rho}(\omega_1, 0) = 0 \quad (\text{B13})$$

and we can write

$$\tilde{G}_{\rho BB}(q; 0, 0) = -\beta \int \frac{d\bar{\omega}}{2\pi} \tilde{G}_{\rho B\rho}(\bar{\omega}, 0) . \quad (\text{B14})$$

In the time domain

$$G_{\rho B\rho}(t_1, t_2, t_3) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} e^{-i\omega_1(t_1-t_3)} e^{-i\omega_2(t_2-t_3)} \tilde{G}_{\rho B\rho}(\omega_1, \omega_2) . \quad (\text{B15})$$

Fourier transforming over t_2 obtains

$$G_{\rho B\rho}(t_1, \omega_2, t_3) = \int \frac{d\omega_1}{2\pi} e^{-i\omega_1(t_1-t_3)} e^{i\omega_2 t_3} \tilde{G}_{\rho B\rho}(\omega_1, \omega_2, -\omega_1 - \omega_2) . \quad (\text{B16})$$

Setting $t_3 = t_1$ and letting $\omega_2 \rightarrow 0$:

$$G_{\rho B\rho}(t_1, 0, t_3) = \int \frac{d\omega_1}{2\pi} \tilde{G}_{\rho B\rho}(\omega_1, 0, -\omega_1) . \quad (\text{B17})$$

Combining Eq.(B14) and (B17) gives

$$\tilde{G}_{\rho BB}(0, 0, 0) = -\beta G_{\rho B\rho}(t_1, 0, t_1) . \quad (\text{B18})$$

When the times of the ρ 's are equal in $G_{\rho B\rho}$ we have

$$G_{\rho B\rho}(q; t_1, t_2, t_1) = \int \frac{d\omega_1}{2\pi} \int \frac{d\omega_2}{2\pi} e^{-i\omega_2(t_2-t_1)} \tilde{G}_{\rho B\rho}(\omega_1, \omega_2) \quad (\text{B19})$$

Introducing $f(t_1) = \delta\rho(q_1 t_1) \delta\rho(q_2, t_1)$ one has $G_{fB}(0)$ satisfies the two-time FDR

$$G_{fB}(0) = -\beta \int \frac{d\bar{\omega}}{2\pi} G_{f\rho}(\bar{\omega}) = -\beta \langle \delta\rho(q_3) f \rangle \quad (\text{B20})$$

$$= -\beta S_{\rho\rho\rho}(q_1, q_2, q_3) \quad (\text{B21})$$

and the static three-point cumulant enters the development. We have then

$$\tilde{G}_{\rho BB}(q; 0, 0) = -\beta G_{fB}(q; 0) = \beta^2 S_{\rho\rho\rho}(q_1, q_2, q_3) \quad . \quad (\text{B22})$$

In terms of the three-point vertex

$$\begin{aligned} \Gamma_{B\rho\rho}(0, 0, 0) &= -\Gamma_{B\rho}(0) \Gamma_{\rho B}(0) \Gamma_{\rho B}(0) G_{\rho BB}(0, 0, 0) \\ &= \frac{\beta^2 S_{\rho\rho\rho}(q_1, q_2, q_3)}{(-\beta S_{\rho\rho}(q_1))(-\beta S_{\rho\rho}(q_2))(-\beta S_{\rho\rho}(q_3))} \\ &= -\beta^{-1} \gamma_{\rho\rho\rho}(q_1, q_2, q_3) \quad . \end{aligned} \quad (\text{B23})$$

which is a result of much use in evaluating the one-loop contribution to the self energy both at single-particle and collective levels. Note that $\gamma_{\rho\rho\rho}$ is a static three-point vertex.

Appendix C: FDR Matrix Propagators

FDR matrix propagators (FDRMP) $A_{\mu\nu}(q, \omega)$ satisfy the following properties:

$$\begin{aligned} A_{\mu\nu}(q, \omega) &= A_{\nu\mu}^*(q, \omega) \\ +i\beta\omega A_{\rho\rho}(q, \omega) &= A_{B\rho}(q, \omega) - A_{\rho B}(q, \omega) \\ A_{\rho B}(q, \omega) &= \int \frac{d\bar{\omega}}{2\pi} \frac{\beta\bar{\omega} A_{\rho\rho}(q, \bar{\omega})}{\omega - \bar{\omega} + i\eta} \quad . \end{aligned} \quad (\text{C1})$$

From this it follows

$$A_{\rho B}(q, 0) = - \int \frac{d\bar{\omega}}{2\pi} \beta A_{\rho\rho}(q, \bar{\omega}) \quad . \quad (\text{C2})$$

Finally the element $A_{BB}(q, \omega) = 0$.

We now prove the following important property of the FDRMP : If $A_{\alpha\beta}(q, \omega)$ and $C_{\alpha\beta}(q, \omega)$ are FDR matrix propagators then

$$D_{\alpha\beta}(q, \omega) = \sum_{\mu\nu} A_{\alpha\mu}(q, \omega) \sigma_{\mu\nu}(q) C_{\nu\beta}(q, \omega) \quad (\text{C3})$$

is also a FDR matrix propagator. The proof is rather direct. Look first at the response channel:

$$\begin{aligned} D_{BB}(q, \omega) &= \sum_{\mu\nu} A_{B\mu}(q, \omega) \sigma_{B\nu}(q) C_{\nu B}(q, \omega) \\ &= A_{B\rho}(q, \omega) \sigma_{\rho\rho}(q) C_{\rho B}(q, \omega) = 0 \quad . \end{aligned} \quad (\text{C4})$$

Consider next the off -diagonal components

$$\begin{aligned} D_{\rho B}(q, \omega) &= A_{\rho B}(q, \omega) \sigma_{B\rho}(q) C_{\rho B}(q, \omega) = A_{\rho B}(q, \omega) V(q) C_{\rho B}(q, \omega) \\ D_{B\rho}(q, \omega) &= A_{B\rho}(q, \omega) V(q) C_{B\rho}(q, \omega) \end{aligned} \quad (\text{C5})$$

It is easy to see that $D_{\rho B}(q, \omega) = D_{B\rho}^*(q, \omega)$. Next consider the diagonal component

$$\begin{aligned} D_{\rho\rho}(q, \omega) &= A_{\rho\rho}(q, \omega) V(q) C_{B\rho}(q, \omega) + A_{\rho B}(q, \omega) V(q) C_{\rho\rho}(q, \omega) \\ &= \frac{V(q)}{i\beta\omega} [(A_{B\rho}(q, \omega) - A_{\rho B}(q, \omega)) C_{B\rho}(q, \omega) + A_{\rho B}(q, \omega) (C_{B\rho}(q, \omega) - C_{\rho B}(q, \omega))] \\ &= \frac{V(q)}{i\beta\omega} [(A_{B\rho}(q, \omega) C_{B\rho}(q, \omega) - A_{\rho B}(q, \omega) C_{\rho B}(q, \omega))] \\ &= \frac{1}{i\beta\omega} [D_{B\rho}(q, \omega) - D_{\rho B}(q, \omega)] \end{aligned} \quad (\text{C6})$$

The above result implies that

$$i\beta\omega D_{\rho\rho}(q, \omega) = D_{B\rho}(q, \omega) - D_{\rho B}(q, \omega) \quad (\text{C7})$$

$$-i\beta\omega D_{\rho\rho}^*(q, \omega) = D_{\rho B}(q, \omega) - D_{B\rho}(q, \omega) \quad (\text{C8})$$

Together Eqs.(C7) and (C8) give $D_{\rho\rho}(q, \omega) = D_{\rho\rho}^*(q, \omega)$.

Let us now consider the dressed propagators respectively denoted as \bar{G} and \tilde{G} . In operator notation \bar{G} and \tilde{G} are respectively defined as $\bar{G} = \mathcal{G}\sigma G$ and $\tilde{G} = \mathcal{G}\sigma G\sigma\mathcal{G}$. Writing out explicitly the matrix forms we obtain for \bar{G} and \tilde{G} the following expressions :

$$\bar{G}_{\alpha\beta}(q, \omega) = \mathcal{G}_{\alpha\mu}(q, \omega) \sigma_{\mu\nu}(q) G_{\nu\beta}(q, \omega) \quad (\text{C9})$$

$$\tilde{G}_{\alpha\beta}(q, \omega) = \mathcal{G}_{\alpha\mu}(q, \omega) \sigma_{\mu\nu}(q) G_{\nu\delta}(q, \omega) \sigma_{\delta\eta}(q) \mathcal{G}_{\eta\beta}(q, \omega) \quad , \quad (\text{C10})$$

From the above theorem then it follows that both \bar{G} and \tilde{G} are FDRMP. This holds since each of the G , \mathcal{G} , and $G^{(0)}$ satisfies the conditions of being a FDRMP.

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- [19] The static Ornstein-Zernike relation connects the radial distribution function to the direct correlation function.
- [20] In different contexts the kinetic kernels have different names. In the field theory context the kernels are typically called self-energies, in the kinetic theory case, where the analysis is in terms of retarded quantities, the kernel is called a memory function, and in the general case it can be called a dynamic direct correlation function.
- [21] There is an exact solution to the Percus-Yevick approximation for hard spheres. See the discussion in Chapter 4 in J.-P. Hansen and I. R. McDonald, *Theory of Simple Liquids*, Third Ed., Academic Press, 2006; N. W. Ashcroft and J. Lekner, Phys. Rev. **145**, 83 (1966).
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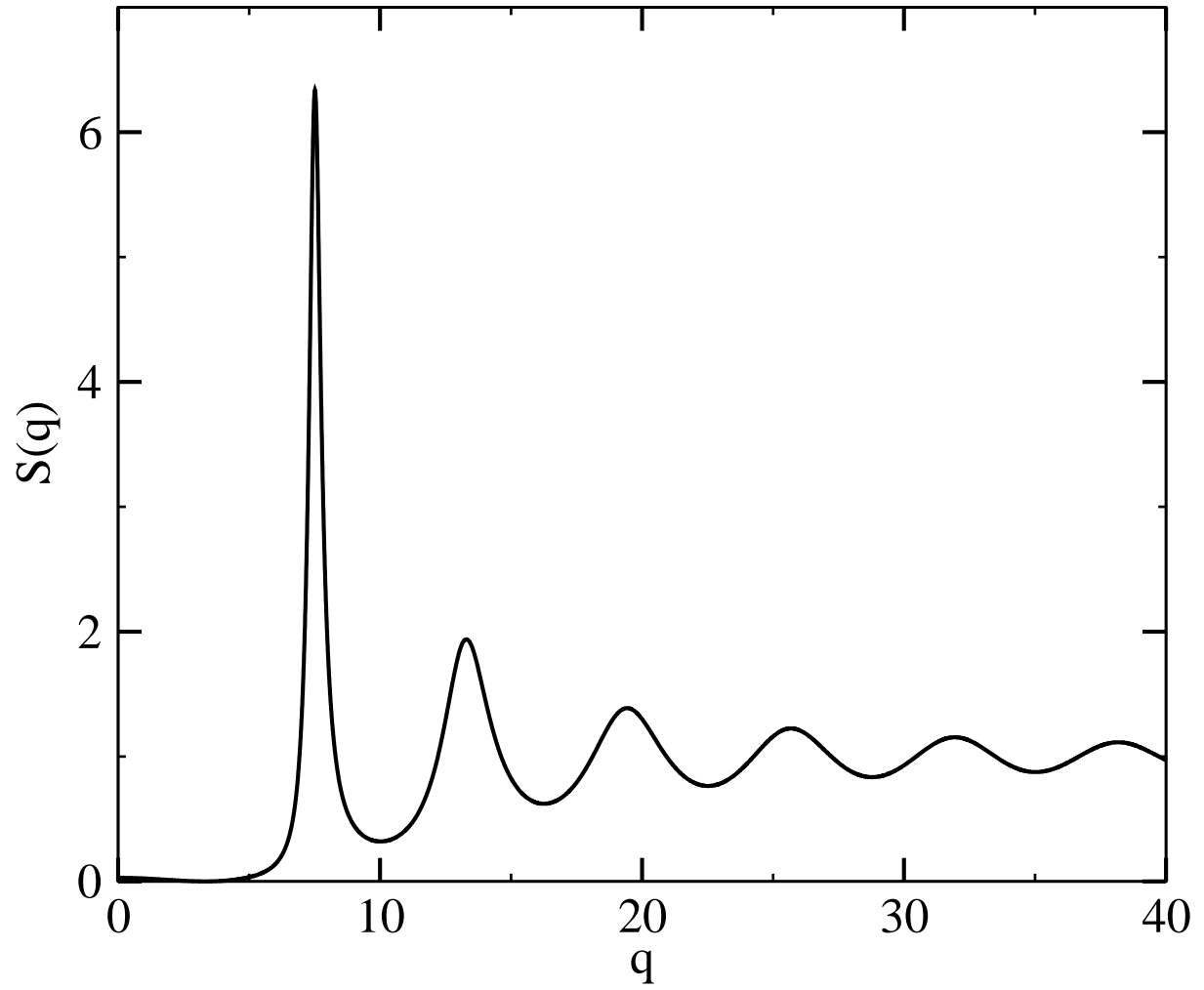


FIG. 1: The static structure factor of the hard sphere liquid at packing fraction $\eta = .62$ as obtained from Percus-Yevick solution with Verlet-Weiss corrections.

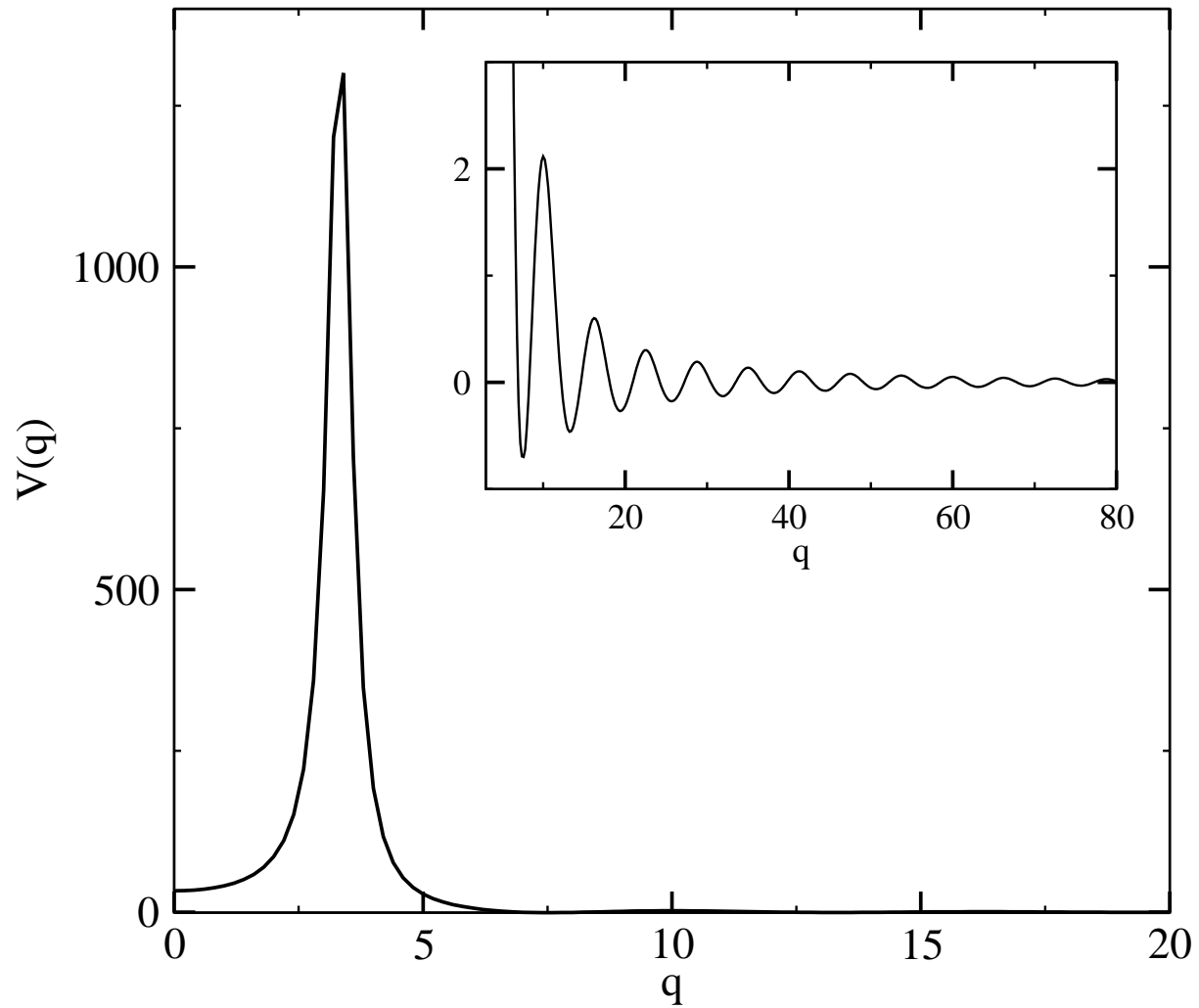


FIG. 2: The renormalized potential $\tilde{V}(q)$ corresponding to the structure factor displayed in Fig. 1. The inset shows the oscillations at large wave vectors on a enlarged scale.

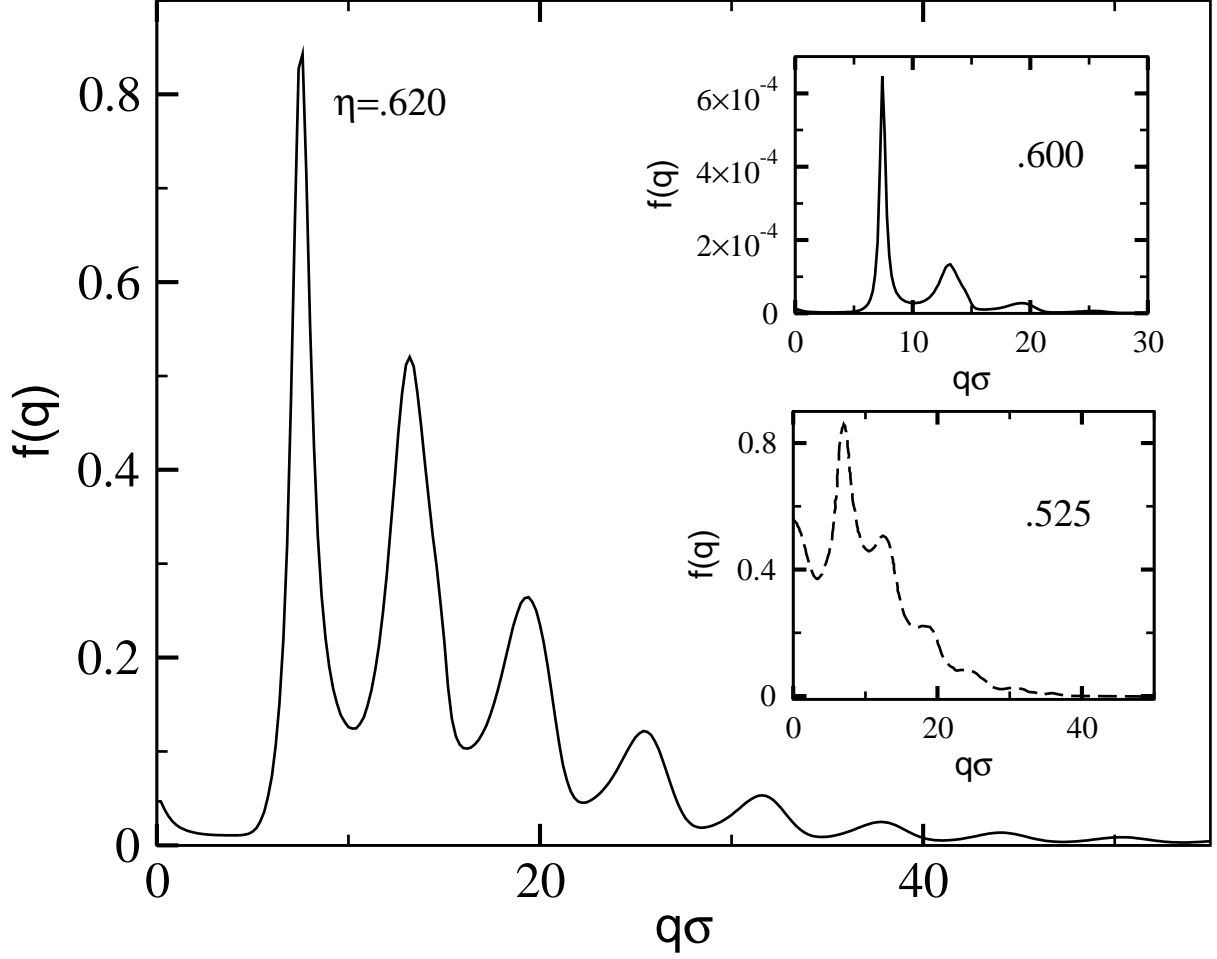


FIG. 3: The non ergodicity parameter $f(q)$ vs. $q\sigma$ at packing fraction $\eta = .62$. The upper inset shows the same quantity at $\eta = .60$ indicating ergodic behavior. The lower inset shows the $f(q)$ vs. $q\sigma$ at the ENE transition point $\eta = .525$ in the earlier MCT of Ref. 10